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THE LIME-SODA SINTER PROCESS FOR RESOURCE RECOVERY FROM FLY ASH — A NEW LOOK

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ABSTRACT

The lime-soda sinter process is one of the earliest and most thoroughly researched and evaluated of the several methods available for resource recovery from fly ash. The principle product, metallurgical grade alumina, is obtained with yields as high as 90% depending upon how much alumina needs to be left in the residue to form acceptable byproduct cement clinker. The process has the advantages of requiring a relatively low sintering temperature (1100-1200°C), using conventional equipment of carbon steel construction, utilizing a variety of calcium and mineralizer raw materials, and producing only a single byproduct consisting of dicalcium silicate that has been shown to be an attractive raw material for the manufacture of portland cement.

An economic feasibility study for a combined facility to produce alumina and cement from the fly ash generated by a 1000 MWe coal-fired power station shows a 10.5% return on average investment. This is increased to 14.2% when a disposal charge of $10/ton of fly ash consumed is credited to the process.

Research has shown that the soda ash can be replaced by coal cleaning refuse or that the soda ash and one-fourth of the limestone can be replaced by FGD sludge with a savings in raw material cost in both cases. The return on average investment becomes 14.5% when the refuse is used and 15.2% when the sludge is used. The return could be increased further if an inexpensive fluxing agent were substituted for the alumina deliberately left in the residue.

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INTRODUCTION

This paper is concerned with recent technical developments and a reassessment of the economic feasibility for the Ames Lime-Soda Sinter Process. The motivation for this reexamination comes in part from the recent EPA study and report to Congress on Wastes from the Combustion of Coal by Electric Utility Power Plants (1). One of three recommendations made in the report calls for utilization as one method for reducing the amount of these wastes that requires disposal. Utilization is endorsed as a management control option that is normally consistent with the goal of protecting human health and the environment. At the same time, it is noted that coal solid waste management is primarily a local responsibility, but also one that is national in scope and requires a national strategy to solve it.

The solid waste that is the best candidate for utilization is fly ash. It is widely available and is becoming increasingly consistent in quality and supply. Resource recovery from the ash can be an attractive utilization option particularly when the entire ash is converted to useful products or byproducts. Recovery of the minerals in the ash is most frequently the primary objective.

Recovery of minerals from fly ash is hindered by the nature of the ash. The vitreous and fused particles of the ash consist of metal silicates, which are difficult to attack. Consequently, work since the mid 1970s at the Ames Laboratory has focused on a high temperature sinter-leach process to break the metal-silica bonds and recover alumina from the clinker.

The sinter treatment of silicate compounds is not new — Seailles patented most of the sinter, extraction and purification steps during the years 1925-1950 (2). The process developed by Seailles was applied to clays, igneous rocks, and coal solid wastes by workers at the Illinois Geological Survey in 1945 (3) and the Bureau of Mines in 1947 (4,5). The method was later investigated in Poland and Hungary for the recovery of alumina and the production of byproduct cement from fly ash (6). The Hungarian work included a pilot plant study but did not show high alumina recoveries from the U.S. fly ashes tested. Research at the Ames Laboratory has identified improved process conditions for U.S. coal fly ashes and has led to development of the Ames Lime-Soda Sinter Process shown in Figure 1.

In the Ames process, ash that contains a significant amount of magnetic material is first fractionated in a magnetic separator to yield an iron-rich portion suitable for use in heavy media or as an iron ore. The remaining fly ash (non-magnetic) is combined with ground limestone and a small amount of soda ash,

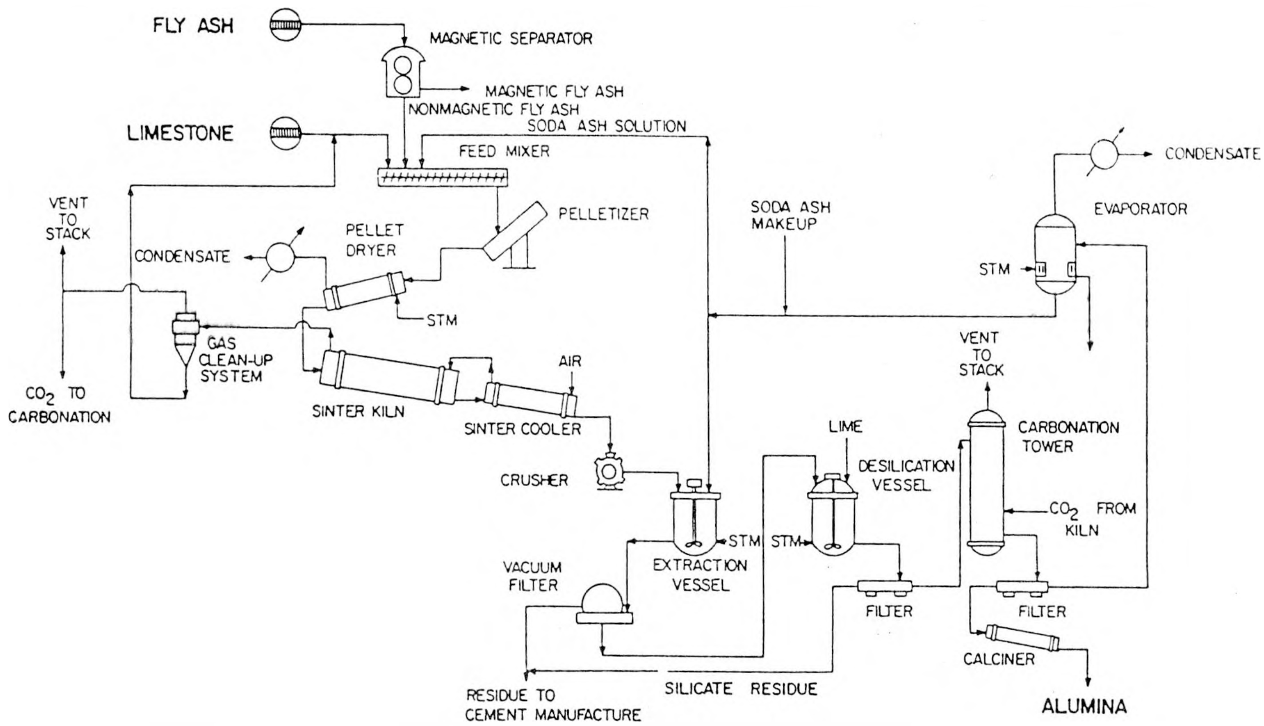


Figure 1. The Ames Lime-Soda Sinter Process for the recovery of Al_2O_3 from power plant fly ash.

mixed, formed into pellets, and sintered. During sintering, soluble sodium and calcium aluminates and insoluble calcium silicate compounds form. The soda ash acts as mineralizer to enhance the reactions taking place and to lower the required sintering temperature by about 150°C to 1100-1200°C. It also results in a significant increase in alumina recovery from 50-60% to 80-90%.

The cooled clinker from the sintering step is ground and leached with a dilute soda ash solution to recover the calcium and sodium aluminates. A small amount of silica that appears in the extract is removed by treatment with lime to form a precipitate of calcium silicate. Treatment of the purified extract with carbon dioxide results in a precipitate of aluminum hydrate, which upon calcination forms metallurgical grade alumina. As will be noted later, the calcium silicate residue from the extraction is an excellent raw material for the manufacture of portland cement.

The Ames Lime-Soda Sinter Process has been investigated through the mini-pilot plant stage (7). The study included the major process variables consisting of the composition of the mixtures sintered, the sintering time and temperature, the composition of the leach solution, and the leaching time and temperature. Perceived advantages for the process include rapid reaction during sintering, rapid leaching and high alumina recoveries, a relatively low sintering temperature, use of conventional equipment of carbon steel construction, ability to use a variety of calcium and mineralizer raw materials, and production of only a single byproduct, primarily calcium disilicate.

USE OF COAL WASTES AS PROCESS MATERIALS

Utilization as an alternative to disposal is also attractive for coal solid wastes other than fly ash. The two wastes found to substitute satisfactorily for regular raw materials in the Lime-Soda Sinter Process are coal cleaning refuse and flue gas desulfurization (FGD) sludge.

Coal Cleaning Refuse

A disadvantage of adding soda ash as a mineralizer in the Lime-Soda Sinter Process is the small but significant increase in raw material cost. In an effort to find an effective, but inexpensive mineralizer, small amounts of coal cleaning refuse were tried with gratifying results (8). Alumina recoveries of over 90% were achieved at a sintering temperature of only 1200°C. The effect on alumina recovery of sintering temperature and time, and of the lime and coal refuse contents in the sinter feed, were investigated. Also of interest were the role and fate of the elements added in the refuse, and the aluminum compounds formed during sintering.

The coal cleaning refuse used was obtained from the Iowa State University coal preparation plant and resulted from the cleaning of a southeastern Iowa high-sulfur coal on a wet concentrating (Deister) table. The chemical compositions of the refuse and of the fly ash used in the investigation are shown in Table 1. The ash is from the burning of a Powder River Basin, Wyoming subbituminous coal.

Aluminum-containing compounds identified in the sintered mixture before soda ash extraction were: calcium aluminoferrite (C_4AF), calcium aluminate ($C_{12}A_7$), calcium aluminosulfate ($C_4A_3\bar{S}$), and gehlenite (C_2AS). Other calcium aluminates such as CA or C_3A may have formed during sintering, but were not in sufficient quantity (less than 1 wt%) to be identified in x-ray diffraction patterns. The four compounds identified accounted for 80-90% of the total aluminum present, depending upon the sintering conditions.

Alumina recoveries of 90% or better were obtained for a refuse addition of 5% by weight, a sintering temperature of 1200°C, a sintering time of 30 minutes, and a CaO/Al_2O_3 ratio of 2.7. Less than 10% of the sulfur added with the refuse left during sintering as SO_2. The amount was a function of the excess sulfur added in the refuse.

Table 1

CHEMICAL COMPOSITION OF THE OTTUMWA SUBBITUMINOUS COAL FLY ASH, THE KCPL LACYGNE FGD SCRUBBER SLUDGE, AND THE COAL CLEANING REFUSE (MINERALIZER) FROM THE ISU COAL PREPARATION PLANT

	Chemical composition, wt%[a]		
Component	Ottumwa fly ash	Scrubber sludge[b]	Coal cleaning refuse
SiO_2	34.40	9.14	3.63
Al_2O_3	19.80	2.62	1.19
Fe_2O_3	5.06	4.22	33.20
CaO	24.90	35.20	4.5
MgO	3.95	0.40	0.16
Na_2O	2.06	0.17	0.02
K_2O	0.36		0.33
S	0.43	8.21	26.70
C		4.08	30.25

[a]Elements other than S and C are reported as oxides but are not necessarily present in that form. All analyses are reported on a dry basis.

[b]About 70 wt% of the sulfur is present as $CaSO_3$; the carbon is present primarily as unreacted limestone, $CaCO_3$.

Flue Gas Desulfurization Sludge

The successful use of coal refuse as a mineralizer led to the investigation of other sulfur-containing wastes. One of these was flue gas desulfurization scrubber sludge, which consists primarily of $CaSO_3$, $CaSO_4$, and unreacted limestone. It was found to serve not only as a mineralizer, but as a partial replacement for the limestone required as a calcium source in the lime-sinter reactions.

The sludge used in the sinter tests was from the LaCygne power station of the Kansas City Power and Light Company, where the fuel was a low-sulfur, subbituminous western coal. The same fly ash was used, that from the Ottumwa, Iowa, power station. The chemical compositions of the sludge, which was of the non-oxidized type, and of the ash are shown in Table 1.

The sintering time and temperature required was found to be the same as when coal refuse was used as the mineralizer. Yields of alumina reached 90% when about 20%

of the CaO required to react with the Al_2O_3 was provided by the sludge. Yields of alumina fell off once this critical level of sulfur addition was reached.

The fraction of the sludge produced at a given power station that could be used in processing the ash from that same station is of interest. When using a fly ash from a typical western coal in the Lime-Soda Sinter Process, about two-thirds ton of limestone is required for each ton of ash consumed. At the power station, about one and one-half tons of wet FGD sludge are produced per ton of fly ash generated. Replacing one-fifth of the limestone required to form aluminate with calcium from the FGD sludge will consume about one-half the sludge produced.

Additional work is required to determine the availability of the CaO in oxidized sludges, which is present primarily as $CaSO_4$, and the fate of the sulfur introduced with the sludges, oxidized or non-oxidized. Preliminary indications are that oxidized sludges may not be as reactive and that in either case, a major fraction of the sulfur remains in the solid residue.

PORTLAND CEMENT FROM THE PROCESS RESIDUE

The Lime-Soda Sinter Process leach residue is of special value in the manufacture of portland cement because the lime present is already combined with silica and only a small amount of supplementary limestone must be added to form alite (C_3S), the principal component in cement. Other advantages are (1) the high specific surface area of the residue, which increases its reactivity and hence the throughput available from a given cement kiln, and (2) the low alumina content, which permits the ready manufacture of sulfate resistant (ASTM Type V) cement. This premium grade cement must be low in C_3A which is difficult to achieve using conventional cement raw materials.

Yet another important advantage from the use of sinter residue is the reduced energy consumption in the kiln per ton of cement produced. This results from less $CaCO_3$ to calcine, less raw mix to bring to sintering temperature, and no bound water to free and evaporate. A thorough theoretical and experimental study by Chesley (10) revealed a 50% reduction in the amount of fuel required by the kiln (a 79% energy saving in the theoretical heat requirements to form cement), and a 24% reduction in kiln throughput.

Cements produced in the laboratory have been found to meet all of the specifications for a Type V product (9), thus a combined lime-soda sinter and cement facility has been used as the basis for the following economic evaluation.

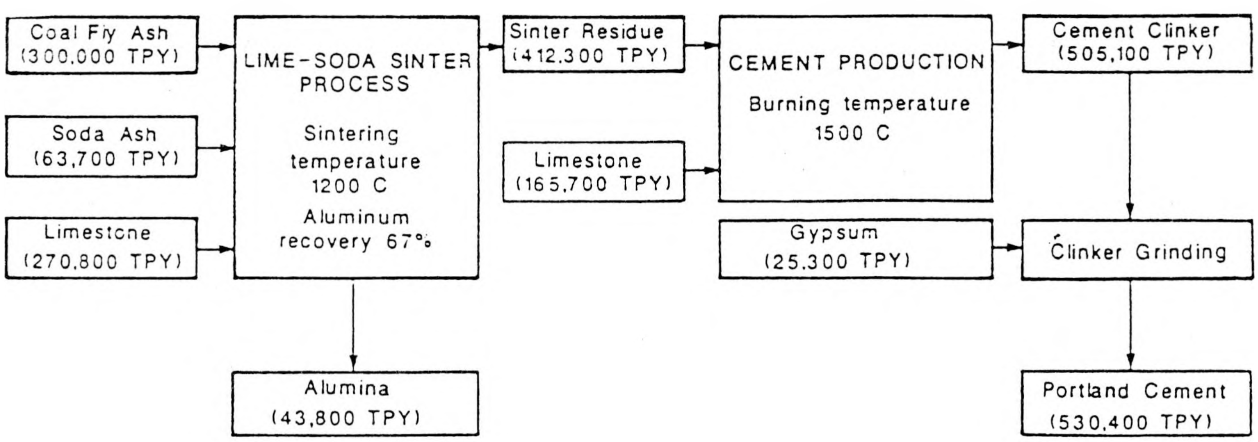
Equipment has been sized to treat 300,000 TPY of fly ash, the amount generated by a 1000 MWe power station burning a low-sulfur western coal.

ECONOMIC FEASIBILITY STUDY

An economic evaluation of a combined Ames Lime-Soda Sinter Process and cement manufacturing facility follows. Included is the estimated capital investment, the annual operating cost, and the potential return on the average investment. In addition, the effect of using coal cleaning refuse and FGD scrubber sludge as raw materials, and of plant size on profitability are examined.

Material Balance

A material balance flowsheet for the Lime-Soda Sinter Process and cement manufacturing facility is shown in Figure 2. A 67% alumina recovery from the fly ash is assumed so that no alumina as flux is added to the raw mix. This results in the production of 43,800 TPY of alumina (including alumina from the ash generated by the combustion of coal to direct-fire the cement kiln) and 530,400 TPY of portland cement. A common capacity for a cement plant is one million TPY which



Coal Fly Ash — Approximate Analysis

	% w
CaO	23.3
SiO_2	38.8
Al_2O_3	18.1
Fe_2O_3	4.8

Sinter Residue — Approximate Analysis

	% w
CaO	55.3
SiO_2	27.3
Al_2O_3	4.4
Fe_2O_3	4.0

Residue-Cement Clinker — Approximate Analysis

	% w		% w
CaO	63.5	C_3S	62.8
SiO_2	22.5	C_2S	17.1
Al_2O_3	3.6	C_4AF	9.9
Fe_2O_3	3.3	C_3A	3.9

Figure 2. Material flowsheet for the lime-soda sinter cement process based on 300,000 TPY coal fly ash.

Table 2

ESTIMATED CAPITAL COST FOR A COMBINED LIME-SODA SINTER PROCESS AND PORTLAND CEMENT MANUFACTURING FACILITY[a]

Item	Capital Costs, $ Millions	
Lime-soda sinter process		
Raw material preparation	4.15	
Crushing, grinding		
mag. sep. and pelleting		
Sintering	9.10	
Leaching	0.85	
Desilication	1.20	
Carbonation and calcination	1.65	
Soda ash recovery	0.80	
Flue gas (CO_2) processing	0.60	
Cement production	45.30	
Total installed equipment cost[b]		63.65
Buildings and yard[c]	6.35	
Plant Utilities[d]	7.65	
Total direct plant costs		77.65
Engineering and supervision	3.90	
Construction expense	8.55	
Total direct and indirect costs		90.10
Contractor's fee	4.50	
Contingency	13.50	
Fixed capital investment		108.10
Working capital	16.20	
Total Capital Cost		124.30

[a]For a power station (1000 MWe) producing 300,000 tons of fly ash per year.

[b]Includes delivered equipment cost plus all foundations and structures, instrumentation, electrical, piping, insulation, painting, and miscellaneous expenses.

[c]Includes buildings, laboratories, shops, roads, but not land.

[d]Includes steam, water, and power distribution, cooling towers, and fire protection equipment.

will result in a lower unit cost than for the smaller plant assumed here. This could be compensated for by processing a larger amount of fly ash.

Capital Costs

The major pieces of equipment required are of standard manufacture and quite similar for the Lime-Soda Sinter Process and for the production of cement. Both operations consist essentially of high temperature solids processing. Equipment requirements in common include grinders, mixers, storage silos, rotary kilns, and hoppers. The Lime-Soda Sinter Process additionally requires magnetic separators, reaction vessels, filters, and tanks.

The capital cost for a combined lime-soda sinter, cement facility to process 300,000 TPY of fly ash has been estimated to be about $124 million as shown in Table 2. A summary of the capital costs for the dry process cement plant to utilize the residue from the alumina facility is shown in Table 3. The cement plant costs are for a conventional plant processing 400,000 TPY clinker (11) rather than 505,100 TPY clinker estimated for the residue-cement plant. This adjustment is required because of the low $CaCO_3$ content in the sinter residue cement raw mix, which results in less energy being consumed per ton of product. Also, the kiln throughput is less per ton of product (estimated at 24%).

Table 3

ESTIMATED CAPITAL COSTS FOR THE DRY-PROCESS RESIDUE-CEMENT PLANT[a]

Department	Capital Cost, $ Millions[b]
Raw storage	1.55
Homogenizing and kiln feed	4.70
Clinkering and cooling	18.40
Clinker storage	5.45
Finish grinding	6.25
Cement storage	9.95
Total	45.30

[a]For a cement plant producing 505,100 TPY of clinker. See Figure 2.

[b]Includes delivered equipment cost except for cement feed grinding equipment plus all foundations and structures, instrumentation, electrical, piping, insulation, painting, and miscellaneous expenses.

Production Costs

Estimated alumina and cement production cost data for four different raw material options are given in Table 4. The options deal with the cost of the fly ash and with replacement of the soda ash and the limestone with coal solid wastes. The raw material prices and utility rates used are detailed in Table 5.

The most significant reduction in production cost (Case 2) results from crediting the process with $10 for each ton of fly ash consumed. This is not unreasonable in view of the disposal costs being paid by utilities today (12). Power plants that dispose of their ash in a dry state experience from $2 to $18 a ton disposal costs. The amount is $5 to $31 per ton for plants that dispose of their ash in ponds.

Significant but smaller reductions in production cost result from the replacement of the soda ash with coal cleaning refuse (Case 3) and from replacement of the soda ash and one-fifth of the limestone with FGD sludge (Case 4). The process is credited with $7 in lieu of disposal costs for each ton of sludge consumed.

Profitability

An estimate of the profitability for each of the four options is shown in Table 6. The return on average investment varies from 10.5% for the base case to 15.2% in Case 4 where credit is taken for the disposal of the fly ash and the FGD sludge, and sludge is used to replace the soda ash and one-fifth of the limestone. At the state of development of the technology involved, these returns are modest but encouraging.

A key item in the profitability analysis is the selling price for alumina. The current published price of $418/ton used in Table 6 is toward the high end of the price range experienced in recent years. The price is influenced by international market factors that are difficult to predict.

The profitability could be improved if another form of flux were employed other than the alumina left in the residue from the lime-soda sinter process. If the alumina recovery were increased from 67% to 85%, an additional $4.1 million in annual sales could be realized. A possible flux material would be an inexpensive alumino silicate clay.

Table 4

ANNUAL PRODUCTION COSTS FOR A COMBINED LIME-SODA SINTER PROCESS AND PORTLAND CEMENT MANUFACTURING FACILITY[a]

Item	Product Cost, $ Millions[b]			
	Case 1	Case 2	Case 3	Case 4
Direct Cost				
Raw Materials:				
Coal fly ash	1.80	(3.00)	(3.00)	(3.00)
Limestone	2.60	2.60	2.60	2.20
FGD sludge	NA	NA	NA	(0.60)
Soda ash	0.70	0.70	NA	NA
Coal cleaning refuse	NA	NA	0.15	0.15
Gypsum	0.55	0.55	0.55	0.55
Total Raw Materials	5.65	0.85	0.30	(0.70)
Utilities:				
Coal	3.40	3.40	3.45	3.50
Electric power	3.20	3.20	3.25	3.30
Steam	0.40	0.40	0.40	0.40
Water	0.30	0.30	0.30	0.30
Total Utilities	7.30	7.30	7.40	7.50
Direct Labor:				
Labor	2.70	2.70	2.75	2.80
Supervision	1.25	1.25	1.25	1.25
Total Labor	3.95	3.95	4.00	4.05
Other Direct Costs:				
Plant maintenance	2.60	2.60	2.65	2.70
Operating supplies	0.40	0.40	0.40	0.40
Lab charges	0.25	0.25	0.25	0.25
Total Others	3.25	3.25	3.30	3.35
Total Direct Cost	20.15	15.35	15.00	14.20
Fixed Cost				
Taxes and insurance	4.25	4.25	4.25	4.25
Depreciation	5.55	5.55	5.55	5.55
Total Fixed Cost	9.80	9.80	9.80	9.80
Plant Overhead	4.20	4.20	4.20	4.20
General Costs				
Administration	1.05	1.05	1.05	1.05
Sales	1.60	1.60	1.60	1.60
R and D	1.20	1.20	1.20	1.20
Total General Costs	3.85	3.85	3.85	3.85
Total Production Cost	38.00	33.20	32.85	32.05

[a]Combined facility produces 43,800 TPY alumina and 530,400 TPY portland cement; operational year of 330 days.

[b]See footnotes on Table 5 for description of cases.

Table 5

COMBINATIONS OF RAW MATERIAL AND UTILITY COSTS

Item	Costs			
	Case 1[a]	Case 2[b]	Case 3[c]	Case 4[d]
Coal fly ash, $/ton	6	(10)	(10)	(10)
Limestone, $/ton	6	6	6	6
FGD sludge, $/ton	NA	NA	NA	(7)
Soda ash, $/ton	84	84	NA	NA
Coal cleaning refuse, $/ton	NA	NA	5	NA
Sinter residue, $/ton	0	0	0	0
Gypsum, $/ton	22	22	22	22
Coal, $/ton	30	30	30	30
Electric power, $/kW-hr	0.04	0.04	0.04	0.04
Steam, $/ton	0.50	0.50	0.50	0.50
Water, $/1000 gal	0.90	0.90	0.90	0.90

[a]Base case. Charge of $6.00/ton for the fly ash. No sludge or refuse used.

[b]Disposal credit of $10.00/ton for the fly ash. No sludge or refuse used.

[c]Disposal credit of $10.00/ton for the fly ash. Soda ash replaced by cleaning refuse. No sludge used.

[d]Disposal credit of $10.00/ton for the fly ash. Sludge used to replace soda ash and one-fifth of the limestone.

Effect of Plant Size

The average cement plant kiln size in the U.S. is about 500,000 TPY clinker, although kilns range up to twice that size. For this reason, the effect of larger plant sizes on profitability has been investigated.

Table 7 contains the profitability analysis for five levels of plant size using Case 2 where disposal credit is taken for the fly ash but no sludge or cleaning refuse is used. A facility processing 600,000 TPY fly ash shows an attractive return on average investment of 20.3% and requires a cement kiln with a capacity of less than 1 million TPY.

Table 6

PROFITABILITY FOR THE PRODUCTION OF ALUMINA AND CEMENT FROM THE COAL FLY ASH GENERATED FROM A 1000 MWe POWER STATION

Item	$ Millions per Year[a]			
	Case 1	Case 2	Case 3	Case 4
Total sales:				
Alumina ($418 per ton)	$18.30	$18.30	$18.30	$18.30
Cement ($63 per ton)	33.40	33.40	33.40	33.40
	51.70	51.70	51.70	51.70
Production cost	38.00	33.20	32.85	32.05
Profit before taxes	12.70	18.50	18.85	19.65
Federal income tax, 46%	6.30	8.50	8.50	9.00
Net profit after taxes	7.40	10.00	10.15	10.65
Depreciation	5.55	5.55	5.55	5.55
Cash flow	12.95	15.55	15.70	16.20
Payback time, years	9.6	8.0	7.9	7.7
Return on average investment, %	10.5	14.2	14.5	15.2

[a] See footnotes on Table 5 for description of cases.

SUMMARY

Work at the Ames Laboratory has at its goal the transformation of fly ash into our country's sixth most abundant resource by developing a process for economically extracting alumina from the ash and using the byproduct residue for the manufacture of portland cement. It has resulted in the Ames Lime-Soda Sinter Process, which yields metallurgical grade alumina and a dicalcium silicate residue ideal for the manufacture of portland cement. An estimate of a combined alumina/cement facility to process the fly ash from a 1,000 MWe power station shows a return of 14.2% when a disposal credit of $10/ton is taken for the ash consumed. This return is increased with plant size and when the soda ash and a portion of the limestone required are replaced by coal solid wastes other than fly ash.

Table 7

PROFITABILITY FOR PRODUCTION OF ALUMINA AND CEMENT AS A FUNCTION OF PLANT SIZE[a]

Item	Amount of Fly Ash Generated, 1000 TPY				
	200	300	400	500	600
Total Sales:					
Alumina ($418 per ton)	$12.20	$18.30	$24.40	$30.50	$36.60
Cement ($63 per ton)	22.30	33.40	44.55	55.70	66.85
	34.50	51.70	68.95	86.20	103.45
Production Cost	23.70	33.20	42.20	51.80	59.30
Profit before taxes	10.80	18.50	26.75	34.40	44.15
Federal income tax, 46%	5.00	8.50	12.30	15.80	20.30
Net profit after taxes	5.80	10.00	14.45	18.60	23.85
Depreciation	4.10	5.55	6.80	8.10	9.30
Cash flow	9.90	15.55	21.25	26.70	33.11
Payback time, years	9.3	8.0	7.2	6.8	6.3
Return on average investment, %	11.2	14.2	16.8	18.1	20.3

[a]For Case 2. Disposal credit of $10/ton for the fly ash. No sludge or refuse used.

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