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PHYSICAL AND PHYSICOCHEMICAL REMOVAL OF SULFUR FROM COAL

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I. INTRODUCTION

The development of improved coal cleaning methods has been underway at Iowa State University since 1974 when the Iowa Coal Project was established with funds provided by the Iowa Legislature and administered by the Energy and Mineral Resources Research Institute (EMRRI). The primary goals of the effort center on:

1) Demonstration of existing coal cleaning techniques in optimum circuit configurations so as to permit reasonable estimates of their cost and effectiveness.
2) Development and demonstration of new coal cleaning techniques to be utilized in support of existing technologies and to minimize the environmental impact of coal cleaning and utilization.
3) Development of supporting programs in coal characterization and analytical technology for major, minor and trace element determinations and to better understand the nature of coal and its associated minerals such that rational cleaning processes can be developed.

When this research effort was launched, a dual attack was made on high sulfur coal. One approach was the construction of a coal preparation plant capable of demonstrating the performance of selected existing methods for cleaning high sulfur coal which would allow for an assessment of the economics of the processes. A logical selection process chose as the first methods selected for demonstration and evaluation those commercial methods which seemed to have the highest benefit/cost ratio. However, these were gravity separation methods which only removed coarse refuse. Since most high sulfur coal also contains finely disseminated pyrites, a second approach was to screen a number of promising but largely undeveloped methods for removing impurities, to select several methods for further development, and to proceed in developing these methods. The separation methods selected for development include those based on froth flotation, oil agglomeration, hydrocyclones, heavy media cyclones, and high gradient magnetic separation. All of these methods are designed for cleaning fine size coal which is the size that must be cleaned if finely disseminated pyrites are to be liberated and removed and physical coal cleaning is to become effective in meeting some of the various existing and proposed air quality standards. The development of these methods has been uneven since funds were limited. While methods based on froth flotation and oil agglomeration have received extensive bench scale testing and laboratory development leading to the recent installation of a 500-1000 lb/per hour process demonstration unit at the Ames coal research preparation plant, the other methods have received limited attention. However, since hydrocyclones (installed June, 1978) and heavy media cyclones (to be installed Spring 1979) do not lend themselves to small scale systems, these devices have been or are being incorporated into the Ames coal preparation plant and will be evaluated on a demonstration or semi-industrial plant scale.

To improve the separation of coal and pyrites achieved by either froth flotation or oil agglomeration, research has focused on chemical pretreatment of coal fines to enhance the difference in surface properties of the two components. In addition, various combinations of gravity separation, froth flotation, oil agglomeration, and comminution methods have been tested on a bench or demonstration scale to determine what extent these methods complement each other.

II. IOWA STATE UNIVERSITY DEMONSTRATION MINE NO. 1

Before describing our preparation plant, I would like to mention another aspect of our project - the source of some of the nations finest 8% sulfur coal.

A demonstration mine was established on a 40 acre site to determine the potential for reclaiming surfaced mined land for row crop production. Topsoil, nonacid overburden and shale were stockpiled separately during the mining operation and returned in their original layers and re-contoured into bench terraces. (Figure 1)

Originally the site was suitable only for pasture but now contains approximately 25 acres of land suitable for row-crop farming. Studies are being continued to produce optimal conditions for agricultural production.

III. AMES RESEARCH COAL PREPARATION PLANT

A coal preparation plant has been built on the campus of Iowa State University to demonstrate various methods of cleaning coal on a larger scale.

A building to house the plant (Figure 2) and the first section of the plant to clean coarse and medium size coal were completed in 1976. This section included a primary variable speed crusher, heavy media separator, wet concentration table, size separation and dewatering screens, and materials handling equipment. Hydrocyclones for cleaning fine size coal and disc filters for dewatering fines were added in June, 1978; heavy media cyclones for cleaning medium size coal will be added in 1979. All of this equipment is of a semi-industrial scale (approximately 70 TPH) and was funded by the State of Iowa. Pilot plant circuitry to demonstrate our modified froth flotation and oil agglomeration methods of beneficiating fine size coal was installed in 1978 using funds provided by the Fossil Energy Division of the U. S. Department of Energy. A pelletizing circuit was also included at that time. (Figure 3)

The present plant utilizes three processes to separate coal from its impurities. The coal is received as mined and is sized in an impact mill. A vibrating grizzly allows the minus 1½ inch to bypass the crusher. The crushed coal is held in a surge hopper and metered onto the raw coal conveyor. A separating screen pre-wets the coal and separates the plus 3/8" material from the minus 3/8" material.

The 1½" x 3/8" coal is then fed to a cone-shaped heavy media vessel. The clean coal is swept around the surface of the cone and flows over a weir to a scalping screen where the media is returned to the vessel. The coal proceeds across a vibrating screen where any remaining media is washed from the coal and is delivered to the clean coal conveyor and onto a stockpile.

The refuse which has sunk to the bottom of the cone is pumped to a scalping screen to remove the media and then across a vibrating screen where it is rinsed and ultimately delivered to a refuse stockpile.

The 3/8" x 0 coal is delivered from the separating screen to a double-deck concentration table. The coal is washed across the table to a clean coal launder while the refuse passes over the end of the table to the refuse launder. Both streams then pass over sieve bends and onto a parallel bar vibrating screen for dewatering.

The plus 48 mesh cleaned coal and refuse pass onto their respective conveyors while the minus 48 mesh material, along with the process water flows to a fine coal circuit.

The first hydrocyclone is adjusted to permit only clean coal in the overflow while the underflow contains all of the refuse and some coal. The second hydrocyclone increases the accuracy of separation by discharging only refuse through the underflow while permitting only a minimal amount of refuse to be discharged with the coal in the overflow.

Overflow streams, containing clean coal from both hydrocyclones, is thickened in dewatering cyclones and proceeds to a vacuum disc filter for further water removal.

Underflow streams from the hydrocyclones are fed directly to a separate vacuum disc filter for water removal. Dewatered clean coal and refuse are delivered to the output conveyors.

All process water streams eventually flow through a mechanical clarifier where solids are removed with the aid of a flocculant. The water is then completely recycled with only enough makeup to replace the water lost with the plant product.

A heavy media cyclone circuit will be added in 1979 and will be used as an alternative to the wet concentrating table for cleaning medium size coal. The addition of the circuit will not affect other principal features of the plant, but will increase our evaluation capability.

The main plant has been used to demonstrate cleaning of large samples (1000 T) of coal from seven Iowa mines on an industrial scale and to process 40,000 tons of coal from the Iowa State University Demonstration Mine. The samples from the different mines contained from 2.5 to 8.7% total sulfur and from 11.6 to 20.0% ash. (Figure 4) As a result of processing in the plant, the total sulfur content was reduced an average of 35% with a range of 24 to 45% and the ash content was reduced an average of 45% with a range of 34 to 57% for the series of coals. Moreover the pyritic sulfur content was reduced an average of 52% with a range of 37% to 70%. The average weight yield was 75% with a range of 66 to 80% and the average calorific yield was 84% with a range of 74 to 96%. Since these results were obtained before they hydrocyclones and filters were installed, none of the -48 mesh coal was recovered and therefore the yields were lower than would be obtained with the present equipment.

IV. FROTH FLOTATION AND OIL AGGLOMERATION DEMONSTRATION UNIT

A unit for demonstrating the froth flotation and oil agglomeration methods of cleaning fine-size coal has been installed in the Ames coal preparation facility. (Figure 5 & 6) This unit includes equipment for grinding and chemically pretreating 1000 lb. batches of coal and for continuously beneficiating the pretreated coal by froth flotation or oil agglomeration at a rate of 100-200 lb/hour. The circuit also includes a means for pelletizing the beneficiated coal.

Coal fines from the Ames coal preparation plant are placed as an aqueous slurry in either of two agitated tanks which serve for both storage and chemical pretreatment. For the pretreatment step, an alkali is added to the coal slurry which is then heated to the required temperature. Air is introduced next to oxidize the surface of the pyrite particles after which the slurry is cooled to a set temperature for the subsequent separation steps. If a finer particle size is desired, the coal is ground with a ball mill before applying the chemical treatment. The ball mill circuit includes cyclones for both thickening the pulp supplied to the mill and classifying the particles according to size. Consequently, only the coarser particles enter the ball mill.

After the feed has been adequately ground and/or pretreated, it is pumped to either a bank of froth flotation cells or the first stage of the oil agglomeration system. If the feed is directed to the bank of flotation cells, a frothing agent is added and the coal is floated and removed in the froth while the refuse is removed in the underflow. The float product is either filtered to recover the coal or placed in a storage tank to await further treatment.

Either coal fines cleaned by froth flotation or coal fines which have only been chemically pretreated can be oil agglomerated. A slurry of these fines is delivered to the first stage of a two stage agglomeration system. Fuel oil is added and microagglomerates are produced by high shear mixing. The suspension of microagglomerates is conducted to a vibrating screen for dewatering and desliming. The microagglomerates are resuspended in fresh water in the second stage where less vigorous agitation promotes the coalescence and growth of large agglomerates. The suspension is then dewatered on another vibrating screen. The agglomerated coal can either be recovered at this point or conveyed to an inclined rotating disc pelletizer for further size enlargement.

Since construction of this unit was completed only recently, a program of demonstration runs is just getting underway. The results will be evaluated in terms of the recovery of combustible matter, the sulfur, ash, and moisture content of the product, and its physical properties. The effects of important parameters such as residence time, slurry concentration, reagent concentration, and temperature will be applied to several different representative coals from various regions of the country.

V. HIGH GRADIENT MAGNETIC SEPARATION

"High gradient magnetic separation" process is being investigated to determine its feasibility in cleaning high sulfur coal. In this process fine coal in a water slurry is passed through a magnetic field in which a mesh of ferromagnetic material (stainless steel wool) has been placed. The refuse, which has different magnetic properties than the coal, is attracted to the mesh thereby performing the separation. When the mesh is loaded with refuse either the magnet is turned off or the mesh is removed from the field allowing the refuse to be flushed away. This process can be enhanced by seeding the raw coal to increase the magnetic susceptibility of the refuse. This project is presently in the planning stage.

Presently, laboratory studies are underway to determine the relative merits of induction heating, microwave heating, and chemical treatment to alter magnetic properties.

VI. SLAGGING AND FOULING CHARACTERISTICS OF RAW VS CLEANED COALS

To determine the relative effect cleaning coal has on boiler operation, the ashing properties of four coals were determined before and after cleaning.

Although the number of coals was limited, the results show a significant benefit through cleaning. (Grieve, 1978)

A complete chemical analysis was performed on the ash from four coals which had widely varying sulfur content to determine the base content, acid content, base/acid ratio, silica/alumina ratio, silica value, iron value and dolomite percentage. The fouling index was computed by multiplying the base/acid ratio with the percent dry sodium and the slagging index by multiplying the base/acid ratio with the percent dry sulfur. These indexes were compared to indicate the effect cleaning coal has on boiler operation. (Attig and Dunzy 69)

Of the four coals selected, the reduction of the slagging index was least in the lowest sulfur coal and dramatically reduced in the other three. (Figure 7) A lesser reduction in the fouling index was also indicated.

The performance observed while burning these coals in the power plant correlates to the calculated values of fouling and slagging indexes.

VII. RELATED PROJECTS

Several projects are also being pursued which relate either directly or indirectly with the utilization of coal as an energy source.

An on-line nuclear sulfur and ash monitoring device is being developed to continuously measure the total ash and the total sulfur in a moving coal stream while at the same time being insensitive to the moisture level.

An on-line x-ray diffraction technique has shown that monitoring of all forms of inorganic sulfur can be continuously monitored along with other selected minerals. This device is also insensitve to moisture levels in the measured stream.

Both the nuclear and the x-ray diffraction techniques give almost instantaneous measurements (approximately 30 seconds).

A process for pelleting fine coal is being investigated with present results showing that much less pressureis required than for conventional briquetting and that higher moisture levels can be accommodated to a degree which may eliminate the customary expensive step of thermal drying.

Methods for separating fly ash into components which may be commercially utilized for the production of aluminum, iron and other metals while removing undesirable elements from the remaining rejects are being investigated.

Nuclear magnetic resonance studies of coal are being pursued to aid in characterization of the basic structure of coal.

Analysis of major, minor and trace elements in high BTU coal gas and effluents resulting from refuse derived fuels are being done using plasma fluorescence spectroscopy and other methods.

VIII. CONCLUSION

Using conventional equipment, physical coal cleaning has demonstrated the removal of 24 to 45% of the sulfur in several high sulfur coals. More intensive physical cleaning, with a smaller size consist, indicates the possibility of increasing the inorganic sulfur removal up to 88%. Research plans include beneficiation tests of various medium and high sulfur coals not previously tested with present equipment and alternate methods.

Related investigations should lead to a better understanding of the basic nature of coal, reclamation of post-combustion products, and improved analysis of process streams both in physical coal cleaning plants and in coal conversion plants.

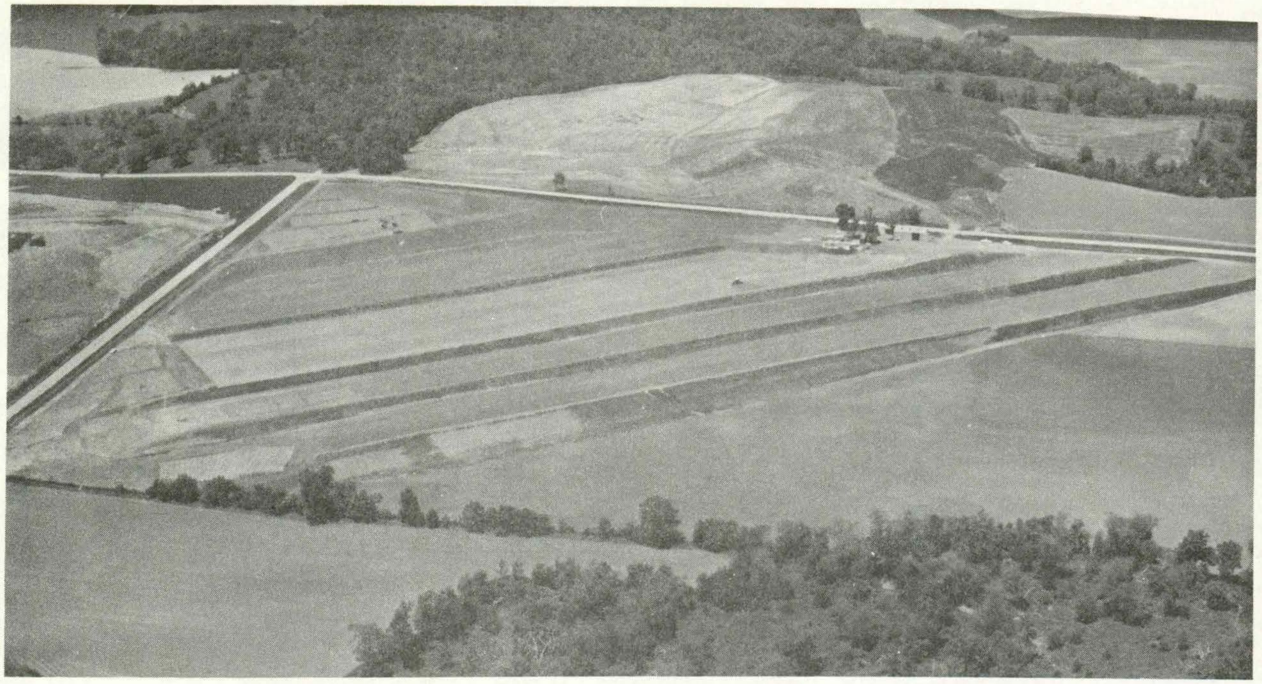
REFERENCES

Attig, R. C., and Duzy, A. F., "Coal Ash Deposition Studies and Applications to Boiler Design," The Babcock & Wilson Company, Alliance, Ohio, Proceedings of the American Power Conference, Vol. 31, 1969, Pages 290 to 299.

Cavallaro, R. A., and Deurbrouck, A. W., "U.S. Bureau of Mines Report of Investigations RI 8118", 1976, Pages 83 to 91, United States Department of the Interior.

Grieve, Richard A., Chu, Henry, and Fisher, Ray W., "Iowa Coal Project Preliminary Coal Beneficiation Cost Study Progress Report," Energy and Minerals Resources Research Institute, Iowa State University, Ames, Iowa (September, 1976).

Figure 1. Aerial View of Restored ICP Demonstration Mine No. 1 Site and Reclaimed Childers Research Site



Laboratory Research Areas

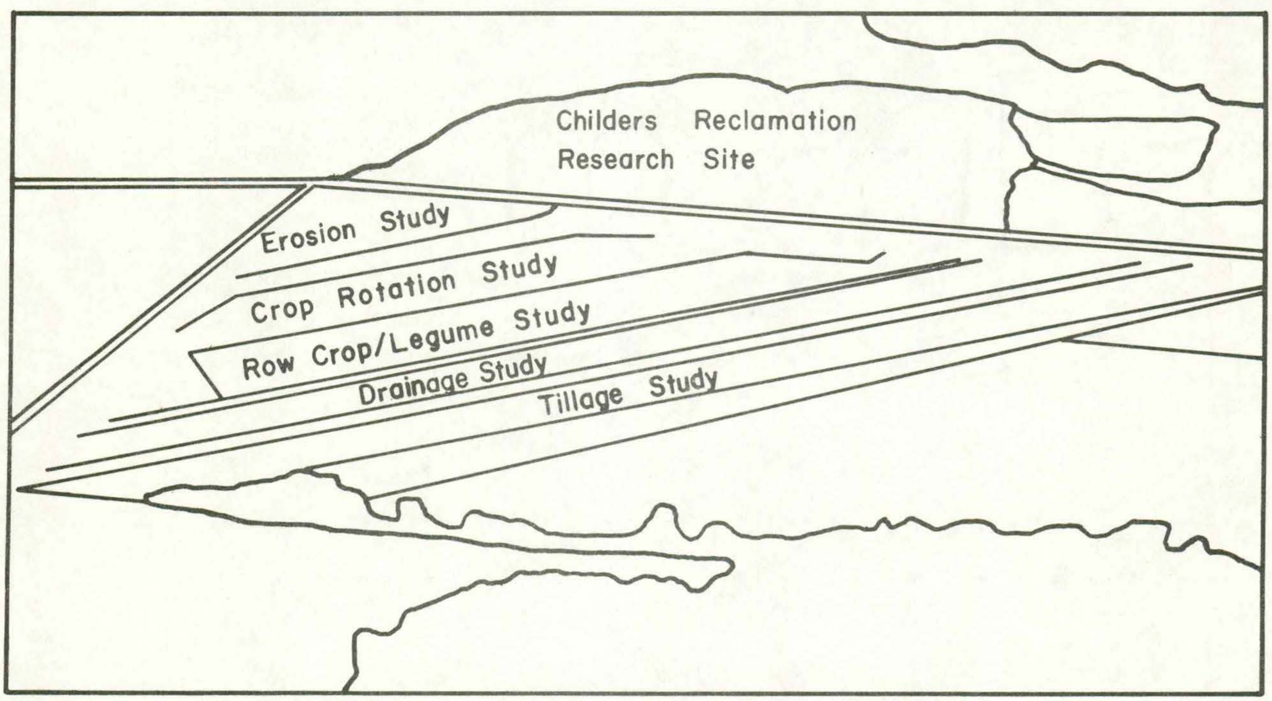


Figure 2. Aerial View of Iowa State University Coal Preparation Plant (white roof) showing close proximity to University Power Plant (Upper R.H. corner of photo)

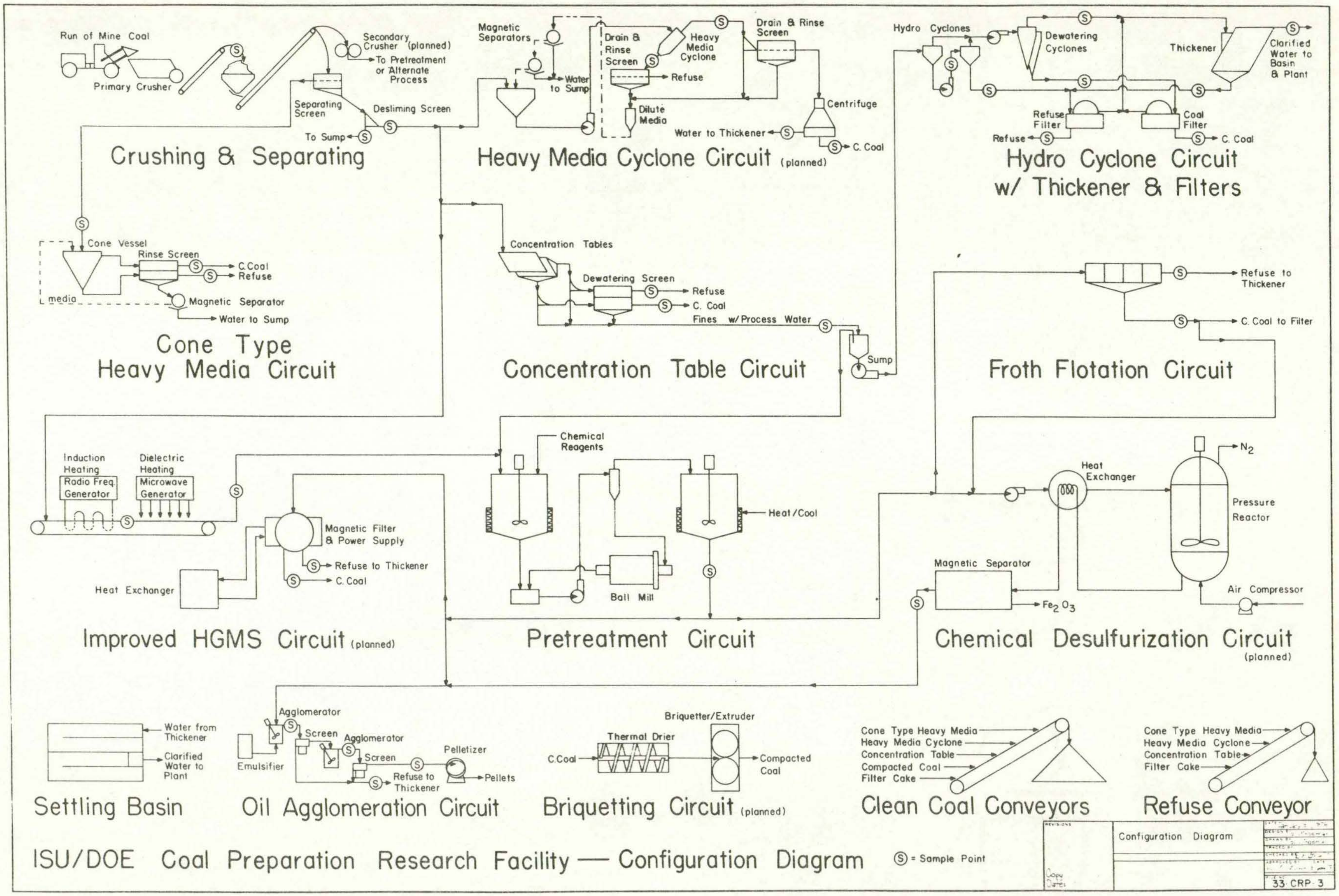


Figure 3. Configuration Diagram

Coal Processed	Specific Gravity	Size		BTU/lb	Total Sulfur (%)	Pyritic Sulfur (%)	Ash (%)	Moisture (%)	Lbs SO_2/MMBTU	Tonnage Yield (%)	BTU Yield (%)	Ash Reduction (%)	Pyritic Sulfur Reduction (%)	Total Sulfur Reduction (%)	SO_2 Reduction (%)
ISU #1	1.5	1½" X 48M	Raw	10,572	6.95	----	16.21	9.24	13.15	80.0	85.7	33.7	----	29.2	34.0
			Clean	11,312	4.91	----	10.73	9.39	8.68						
ICO	1.35	1" X 48M	Raw	10,690	5.48	----	11.55	12.9	10.25	73.6	78.3	39.4	----	40.4	43.9
			Clean	11,724	3.37	----	7.22	10.2	5.75						
Lovilia[1]	1.50	1½" X 48M	Raw	9,839	2.51	1.77	17.16	11.74	5.10	74.9	86.7	46.5	37.0	23.6	33.9
			Clean	11,868	2.0	1.16	9.57	8.0	3.37						
Mich	1.40	1½" X 48M	Raw	10,222	8.74	6.66	24.51	3.26	17.10	72.7	82.9	40.8	55.7	36.3	44.2
			Clean	11,434	5.46	2.89	14.22	5.12	9.55						
Shinn	1.35	1½" X 48M	Raw	10,558	4.56	3.25	15.99	8.65	8.64	66.3	74.0	46.4	44.1	27.0	34.8
			Clean	12,058	3.40	1.86	8.77	6.49	5.63						
Big[1] Ben	1.60	1½" X 48M	Raw	9,368	4.76	3.90	21.34	18.32	10.16	78.5	86.9	48.8	51.3	42.6	48.2
			Clean	10,511	2.50	1.58	9.84	15.57	4.76						
Jude	1.45	1½" X 48M	Raw	8,070	7.84	6.11	29.96	9.04	19.43	70.8	96.4	56.9	70.2	44.8	59.4
			Clean	10,709	4.22	1.77	12.58	11.39	7.88						
Average of all Coals Tested	1.40	1½" X 48M	Raw	9,903	5.83	4.34	19.53	10.45	11.98	73.8	84.4	44.6	51.7	34.8	42.6
			Clean	11,374	3.69	1.85	10.42	9.45	6.52						
						B.O.M. Comparative Data (1) 1½" X 100M @ 1.40 S.G.				72.3	78.8	46.5	59.5	38.0	43.3

[1]Deep Mine. All others surface mines.
[2]Losses in yield computations include fines (-48M) losses.

All proximate analysis - Air dried basis.
All reduction factors - Moisture free basis.

Figure 4. Iowa Coals Processed at Iowa Coal Project Preparation Plant
Data Represents Best Run Made on Each Coal

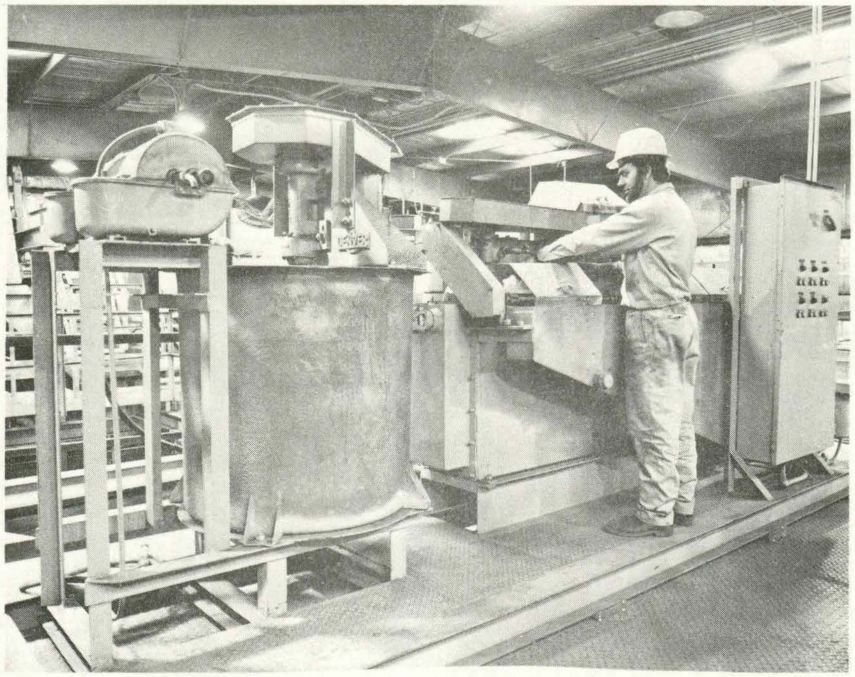


Figure 5. The recovery of fine coal will be demonstrated by this unit consisting of reagent feeder, conditioning tank, and bank of four froth flotation cells.

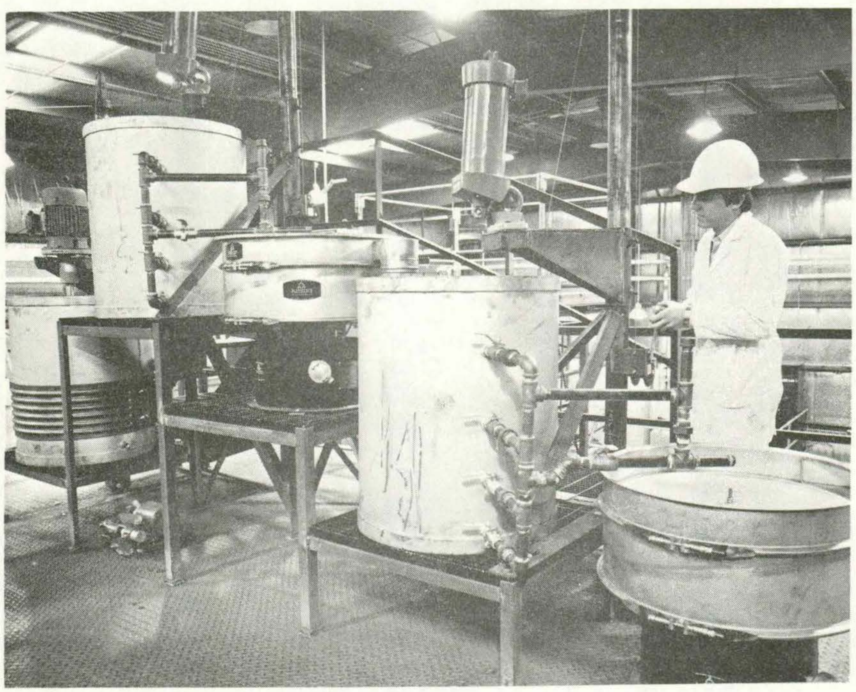


Figure 6. The oil agglomeration and recovery of fine coal will be demonstrated by this system of agitated tanks and dewatering screens.

	ISU (strip mined)		Lovilia (deep mined)		MICH (strip mined)		ICO (strip mined)	
[1]Coal Proximate Analysis	RAW	CLEANED	RAW	CLEANED	RAW	CLEANED	RAW	CLEANED
% Sulfur	11.52	5.42	2.23	2.17	8.74	5.46	7.10	4.94
% Ash	17.86	11.84	12.06	10.40	24.51	14.22	18.28	8.94
BTU	11,384	12,484	12,534	12,901	10,222	11,434	10,896	11,794
Specific Gravity								
Ash Analysis								
[2]Ash Fusion Temp (F°)	2,087	2,110	2,107	2,150	2,037	2,073	2,147	2,153
Base Content	52.63	35.01	40.39	39.35	53.99	46.78	76.10	65.01
Acid Content	34.41	60.61	50.24	49.74	33.17	36.98	14.82	31.16
Base/Acid Ratio	1.53	.58(62%)[3]	.80	.79(1%)[3]	1.63	1.27(22%)[3]	5.13	2.09(59%)[3]
Silica/Alumina Ratio	1.83	1.67	2.03	2.34	2.09	1.95	1.35	2.01
Silica Value	30.39	50.78	47.20	48.66	29.59	35.14	10.11	24.69
Iron Ratio	2.49	4.51	1.48	1.12	3.07	1.51	4.90	8.95
Dolomite Percentage	27.61	17.15	37.39	43.76	24.15	38.26	16.77	9.77
[4]Fouling Index	.20	.09	.29	.33	.09	.02	.13	.03
[5]Slagging Index	17.63	3.14	1.78	1.71	14.72	7.32	38.17	11.24

Fouling Type

Low	Less than 0.2
Medium	0.2 - 0.5
High	0.5 - 1.0
Severe	Greater than 1.0

Slagging Type

Low	Less than 0.6
Medium	0.6 - 2.0
High	2.0 - 2.6
Severe	Greater than 2.6

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1. Air dried basis
2. Average of Initial Deformation, Fusing, and Fluid Temperature under reducing atmosphere conditions
3. Percent reduction from raw coal.
4. (Base/acid ratio) X (%Na)
5. (Base/acid ratio) X (Dry %s)

Figure 7. Comparison of Ashing Properties of Raw vs Beneficiated Coals