



## Project Summary

# Process Improvement Studies on the Battelle Hydrothermal Coal Process

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This report gives results of a study to improve the economic viability of the Battelle Hydrothermal (HT) Coal Process by reducing the costs associated with liquid/solid separation and leachant regeneration.

Laboratory experiments were conducted to evaluate process improvements for (1) separating the spent leachant and residual sodium from the coal product, (2) reducing the moisture content of the coal product, and (3) regenerating the leachant. In addition, coal desulfurization experiments were performed and economic studies were conducted to evaluate the impacts of process improvements on coal desulfurization costs.

Through the use of -20 and -50 (rather than -200) mesh coal and other process modifications, significant process improvements were realized. Separation rates were increased manyfold by adding dispersants. The moisture content of the coal product was lowered to about 40 percent by centrifugation. Sodium was effectively washed from the coal product by saturated lime water. Using countercurrent washing, the optimum washing circuit was composed of four disc filter stages, six belt filter stages to separate spent leachant and sodium from the clean coal, and a centrifuge stage to dewater the coal.

Several regenerates were found to be effective in removing greater than about 85 percent of the total sulfide sulfur from the spent leachant: iron carbonate was the leading candidate, with up to 99 percent removal of the sulfide

sulfur in less than 15 minutes, depending on the Fe/S ratio and source of  $\text{FeCO}_3$ .

Total processing costs (1978 dollars) are estimated to range from \$38/ton of product coal for HT desulfurization of a typical Eastern coal to \$10 for desulfurization of a Western coal. These costs include profit, interest, and tax costs of \$10 and \$4/ton for the Eastern and Western product coals, respectively. Total costs for a combined physical/HT process which cleans high sulfur Eastern coal is estimated to be \$24/ton of product coal.

The process improvements evaluated would provide only marginally lower costs than those for present processes when considering high sulfur coal. However, replacing evaporators in the washing section with reverse osmosis units could potentially reduce costs by \$2 to \$3/ton. Furthermore, a leachant regeneration process similar to the citrate flue gas desulfurization process could reduce costs by \$5 to \$7/ton. Direct combustion of coal without drying in coal/water mixtures could reduce product cost by \$4/ton. These options require further laboratory research for verification.

*This Project Summary was developed by EPA's Air and Energy Engineering Research Laboratory, Research Triangle Park, NC, to announce key findings of the research project that is fully documented in a separate report of the same title (see Project Report ordering information at back).*



## Introduction

### Process Description

The Battelle Hydrothermal (HT) Coal Cleaning Process is a method for desulfurizing coal. The process involves heating an aqueous slurry of coal and a chemical leachant at moderate temperatures and pressures to extract a significant portion of the sulfur and some of the ash. After the reaction step the leachant is washed from the coal and regenerated for recycle to the HT reactor. The process, shown in Figure 1, entails five major processing steps: (1) coal preparation, (2) HT treatment (desulfurization), (3) liquid/solid separation and washing, (4) fuel dewatering and drying, and (5) chemical-leachant regeneration.

Coal preparation entails crushing or grinding of the raw coal, as received from the mine or after washing, to the particle size suitable for desulfurization. In early studies the coal was generally crushed so that 70 percent would pass through a -200 mesh screen.\*

After preparation, the coal is sent to the slurry tank for mixing with the leachant. Alternatively, the coal may be physically beneficiated to remove easily removable ash and pyritic sulfur and then pumped to the slurry tank.

After mixing with the leachant, the coal slurry is pumped continuously through the HT-treatment (desulfurization) segment where it is heated. Portions of the pyritic sulfur, organic sulfur, and other mineral constituents (ash) react with the leachant and are converted to water-soluble forms. The amounts of sulfur and mineral matter which are removed depend on the process conditions (time, temperature, and leachant) and coal properties. The leachant generally used is a mixture of sodium hydroxide (NaOH), calcium hydroxide (Ca(OH)<sub>2</sub>), and water.

The resulting coal-product slurry is passed through a heat exchanger into the product-separation (washing) segment where the desulfurized coal is separated from the spent leachant by a series of filtration and washing operations.

Next, the desulfurized coal is dried (e.g., in a steam jacketed drier) to remove residual water to produce a clean solid fuel.

The spent leachant from the washing segment is regenerated in the leachant-

regeneration segment where the sulfur is also removed. (NOTE: In early process schemes, the sulfur was removed as hydrogen sulfide (H<sub>2</sub>S), using a carbonation step. The H<sub>2</sub>S from this step was converted to elemental sulfur by a Claus or Stretford sulfur-recovery process. The carbonated liquor was filtered to remove solubilized coal and ash values, treated with lime, and again filtered to remove the calcium carbonate precipitate. The calcium carbonate was calcined to produce lime and CO<sub>2</sub> for recycle. The regenerated leachant was concentrated, its composition was adjusted, and it was returned to the process.)

### Project Objectives

Preliminary results of an earlier EPA program—"Combustion of Hydrothermally Treated (HTT) Coals" (Contract 68-02-2119)—indicated that the HTT coals prepared by the Battelle Hydrothermal Coal Process (BHCP) from selected coals are clean solid fuels that can be burned with little or no sulfur emissions control. Much of the coal sulfur which is not removed by the HTT process is tied up with residual sorbent material in the coal. SO<sub>2</sub> concentrations in the flue gases were well below the 1971 New Source Performance Standards (NSPS).

Original project plans called for additional assessment of the combustion characteristics of HTT coal firing. However, cost studies completed as the project was starting indicated that the HTT process probably would not be competitive with flue gas desulfurization unless process improvements were made to reduce costs. Therefore, the objectives of project were changed to:

- Evaluation of methods to reduce the cost of leachant/coal separation and washing, coal dewatering, and leachant regeneration,
- Evaluation of the HTT process performance in desulfurizing three representative coals, and
- Determining the costs of using the improved HTT process.

## Discussion

### Liquid/Solid Separation and Washing

The separation and washing section interacts strongly with the rest of the HTTC process. Hydrothermally treated coal leaving the separation section con-

tains treated coal, moisture, residual treatment chemicals, and spent leachant. The excess moisture must be removed either before or during utilization of the clean coal; the lost chemicals must be made up. The quantity of spent leachant (while dependent on the water/coal ratio used in the reactor, the washwater/coal ratio, and quantity of leachant removed with the product coal) determines the evaporator and regeneration section loads. Makeup of chemicals is determined by the washing efficiency of each separation stage and the number of stages. Washing efficiency is affected by the quantity of residual moisture and chemicals in the HTT coal. Therefore, selecting the optimum separation and washing circuit is a complicated tradeoff of a number of cost-sensitive variables within the separation system (e.g., minimum moisture content of HTT coal obtainable per type of separation equipment, maximum separation rate obtainable per washing stage per type of separation equipment, and minimum sodium removal levels).

### Performance and Cost Trade-offs

In 1976, the cost of HT desulfurizing coal had been calculated at \$31.56/ton exclusive of the cost of coal. The highest costs were those for the washing and separation sections (Table 1). Therefore, an investigation was undertaken to reduce the costs of the liquid/solid (L/S) separation segment of the process.

First the L/S separation costs were separated into seven cost components as summarized in Table 2. Analysis of these costs indicated that the most significant components were moisture penalty, chemical costs, and capital related expenses. Two factors, moisture content of the HTT coal and separation rate, had the greatest effect on the magnitude of these cost components. Therefore, emphasis in this part of the study was directed toward improving the coal/leachant separation rate while achieving good sodium removal from the cleaned coal and reducing the final coal moisture content. The study objective was to obtain an optimum tradeoff between cost and process capabilities, i.e., minimize cost while maintaining acceptable levels of sodium and moisture in the cleaned coal.

Because of the large scale of the planned application and the treated coal characteristics, separation and dewatering by vacuum filtration and centrifugation was selected for intensive study

\*A table for converting English units to the International System of Units is provided at the back of this Summary.

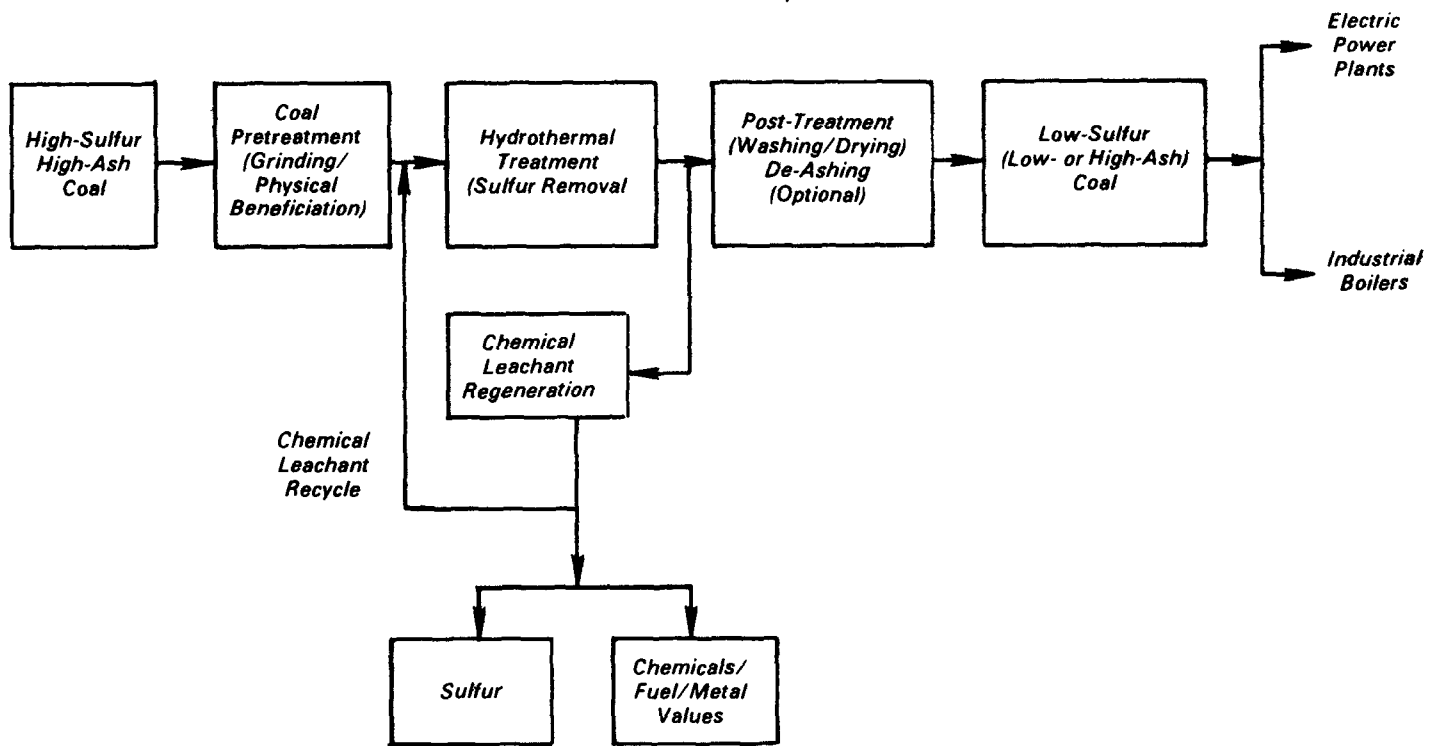


Figure 1. Battelle hydrothermal coal process.

Table 1. Cost of Mixed-Leachant Battelle Hydrothermal Coal Process

Plant Section	Contribution to Price, \$/ton <sup>a</sup> (1976 dollars)
Reactor	5.82
Washing and Separation	9.71
Regeneration	5.25
Sulfur Recovery	3.39
Offsites	7.39
<b>Total</b>	<b>31.56</b>

<sup>a</sup>Processing cost does not include cost of raw coal.

Table 2. Washing and Separation Costs for the Mixed-Leachant Battelle Hydrothermal Coal Process

Cost Components	Contribution to Selling Price, \$/ton <sup>a</sup>
Makeup Chemicals (NaOH)	1.66
Treated Coal	0.97
Utilities	0.26
Direct Labor Related	0.21
Capital Related	1.13
Moisture Penalty	3.83
<b>Contribution to Operating Cost</b>	<b>8.06</b>
Profit, Interest, Income Tax	1.65
<b>Total</b>	<b>9.71<sup>b</sup></b>

<sup>a</sup>First Quarter, 1976 dollars; based on the following treatment conditions: Water/Coal = 2, NaOH/Coal = 0.16, Lime/Coal = 0.05, Reaction Time = 10 min, Reaction Temperature = 527°F, Washwater/Coal = 2

<sup>b</sup>Total processing cost, including \$18.00/ton for raw coal, was \$49.60/ton.

The effects of coal particle size were investigated and the use of filtration aids, surfactants, and oil agglomeration to improve separation rates and final cake moisture were examined. Two types of washing methods were investigated for each of the two separation systems—displacement washing and repump washing.

## Vacuum Filtration

Significant process improvements were realized through the use of coarser coals (-20 and -50 mesh coal, as compared to -200), high NaOH and CaO concentrations, and specialized washing techniques.

## Separation Rate

When the original -20 mesh coal solution was tested, a rate of only 0.008 ton/hr/ft<sup>2</sup> was obtained. Pretreatment testing was first conducted to improve the rate. The use of flocculants resulted in floating of the fines, allowing the coarser material to settle. Consequently, the fines settled on the surface of the cake, resulting in an effective barrier to further dewatering. Dispersants (sodium lauryl sulfate was found most effective) were found to solve this problem by dispersing the fines throughout the cake. Separation rates were increased by a factor of 10, to 0.08 ton/hr/ft<sup>2</sup> at an addition level of 0.5 lb dispersant/ton of coal.

After the initial dispersant addition, the separation rate was found to depend primarily on the degree of washing, increasing after each stage, until it leveled off at >0.6 ton/hr/ft<sup>2</sup>. In addition, the degree of washing also had a moderate effect on the final moisture content of the coal product.

## Moisture Removal

The original separation tests with -20 mesh coal produced a cake with approximately 59 percent moisture. As discussed previously, the use of dispersants to improve the separation rate also improved the moisture removal efficiency during separation. When using dispersants, it was possible to obtain residual moisture contents of about 50 percent with -20 mesh and -50 mesh size particles. A moisture content of about 60 percent was obtained for -60 mesh particles. To obtain satisfactory separation rates and residual moisture contents the product particle size range should be kept above -50 mesh.

Other techniques for reducing coal moisture content included oil agglomeration prior to separation and solvent displacement. The oil agglomeration tests showed that increased separation rates of 1.9 tons/hr/ft<sup>2</sup> could be obtained; however, the moisture content of the clean coal was increased by 1-3 percent. Solvent displacement tests with a mixture of toluene and ethyl alcohol were conducted on a high moisture,

extensively washed filter cake. After filtration the coal filter cake was found to contain the same liquid/solid ratio as the original starting filter cake. Drying tests with the solvent-washed coal showed that drying energy requirements were only half those for water-washed coal. While some of solvent apparently displaced water, the amount of liquid in the solvent displacement filter cake was not substantially different than the original water-washed filter cake.

## Centrifugal Separation

Centrifugal testing data, combined with vendor supplied data, provided the basis for the separation rate and cake moisture content used in the system design. Combined with the data from the filtration section, the centrifugation data were used to design the optimal washing circuit. The results of work on separation rate and moisture removal capability, along with a proposed washing circuit, are summarized separately below.

## Separation Rate

Based on screen-bowl centrifuge equipment used for coal processing of similarly sized coal, a separation rate of about 50 tons dry solids/hr/machine (based on the largest equipment commercially available) has been estimated. No experiment rate tests were conducted (because of the small size of the test equipment), but it is known from theory that the rate is inversely proportional to the liquid viscosity. Therefore, the rate of separation should be improved by higher temperatures. In addition, higher slurry solids concentration should also increase the separation rate.

## Moisture Removal

Tests were conducted to establish the moisture content of the centrifuged HTT coal cake and determine the effect of dispersant additions and oil agglomeration. The tests showed that washed and unwashed HTT coal produced a cake containing about 42 percent moisture. Washing appeared to add little additional moisture to the product cake. The addition of dispersant to the washed coal slurry resulted in cake with about the same moisture content. The dispersant addition, however, did result in greater solids recovery and better centrate clarity. Oil agglomeration, like the dispersant addition, did little to the cake

moisture content, but did improve solids recovery and centrate clarity.

## Washing

The residual sodium in the treated coal must be reduced for economic reasons (for sodium recycle and reuse), as well as combustion (boiler slagging and fouling) considerations.

The washing scheme developed for HTT coal consisted of several counter-current working separation stages in series using repulp washing. Displacement washing methods were found to have low separation rates and high costs.

Tests showed that a saturated lime water wash was clearly superior to washing with water or saturated CO<sub>2</sub> water. Apparently the dissolved calcium in the lime water promoted more effective exchange with the sodium. In fact the bound sodium (sodium not removable by extensive wash) was lowered from about 0.5 percent with water only to about 0.1 percent with lime water. This result was especially significant since it allowed removal to a maximum 0.5 percent total sodium with a reasonable number of washing stages.

Since the process goal is desulfurization, the converted sulfur in the product should be reduced to less than 1.2 lb SO<sub>2</sub>/10<sup>6</sup> Btu by washing. At sufficiently high caustic leachant concentration levels, the sulfur content of the washed coal was consistently brought below the 0.9 percent moisture- and ash-free (MAF) sulfur level. Also, previous studies have shown that the high residual calcium level (>2.6 percent moisture-free (MF) calcium), combined with the residual sodium, led to increased sulfur capture in the ash, making the combustion off-gas even lower in SO<sub>2</sub> than anticipated solely from the coal's sulfur content.

## Combined Separation System

Because of the complex nature of the separation and washing circuit and its interactions with other cost sensitive sections of the BHTC process, a computer program was prepared to investigate the relationships between the total separation and washing costs and the following processing variables: separation equipment, separation rate, cake solids content, washwater/coal ratio, number of washing stages, and residual unbound sodium. Sensitivity studies allowed rapid investigation of the different separation techniques and indicated

where the most significant cost savings could be obtained.

The model studies indicated that a combined system (with a series of filters for washing followed by a final centrifuge stage for dewatering) appeared to be superior to an all-filter system.

### Leachant Regeneration

Spent aqueous caustic soda leachant (utilized in the BHTC Process to remove sulfur and other constituents from coal) contains the sulfur, primarily in sulfide form, that has been extracted from the coal. Work was conducted to develop an improved method for regenerating this spent leachant (i.e., removing sulfur) so the leachant could be recycled to the process.

Previous work at Battelle on the recycle of the NaOH leachant solution had shown that the desulfurizing effectiveness of the leachant decreased as the concentration of sulfide sulfur in the solution increased. The results (see Figure 2) indicated, however, that acceptable desulfurization could be obtained with sulfide concentrations as high as about 0.13 percent (~0.089 lb/cu ft).

Comprehensive review and exploration of sulfur chemistry have revealed only effective leachant desulfurization reactions that involve sulfide sulfur forms. These reactions were of two types: (1) evolution of sulfide in the gas phase as H<sub>2</sub>S, and (2) the precipitation of insoluble sulfides.

Thus, there were two requirements for the leachant desulfurization process: the retention of reacted sulfur in an insoluble sulfide state by avoiding oxidation, and reduction of residual sulfide concentration in the regenerated leachant to a low level.

The candidates initially investigated for regeneration of the spent leachant were:

1. Zinc compounds—zinc oxide and sodium zincate,
2. Iron compounds such as Fe(OH)<sub>2</sub>, Fe(OH)<sub>3</sub>, reduced activated iron oxide, Fe<sub>2</sub>O<sub>3</sub> · H<sub>2</sub>O, FeO(OH), Fe<sub>3</sub>O<sub>4</sub>, elemental iron, hematite, iron carbonyl, water-soluble iron compounds—sodium ferrite, iron nitrate, and iron carbonate,
3. Activated carbon,
4. Electrolysis,
5. Lime, and
6. Copper.

The major approach to the regeneration of leachant was concerned with the use of metallic oxides to remove the sulfide from the spent leachant. Two oxides were studied—iron oxide and zinc oxide. A screening study was conducted to select the more effective, and then the better system was refined to develop a near-optimum set of processing conditions for removing the sulfide sulfur and regenerating the resultant metallic sulfide to obtain the original metallic oxide for recycle.

The metallic oxides do not remove the extracted trace metals. Therefore, buildup of impurities, such as trace metals and solubilized coal, in the regenerated leachant could progressively decrease desulfurization efficiency and contaminate the product coal as the regenerated leachant is recycled. To evaluate this effect, a series of recycle experiments were conducted to determine how many times the regenerated leachant can be recycled and if a bleed stream is needed to prevent contamination of the coal product.

Also, metallic oxides do not remove the oxidized sulfur forms—thiosulfate, sulfite, and sulfate—of sulfur from the spent leachant. These oxidized sulfur forms, which must be removed from the leachant before it is reused for coal desulfurization, are believed to be produced during the desulfurization process or on exposure of the spent leachant to atmospheric oxygen. As part of this subtask, efforts were directed toward controlling these oxidized sulfur forms. Several approaches were studied:

1. Maintaining the spent leachant under a nonoxidizing atmosphere at all times,
2. Reduction of the oxidized sulfur with, for example, metals such as iron and/or zinc and gaseous hydrogen during the regeneration of the spent leachant and/or during the desulfurization operation.

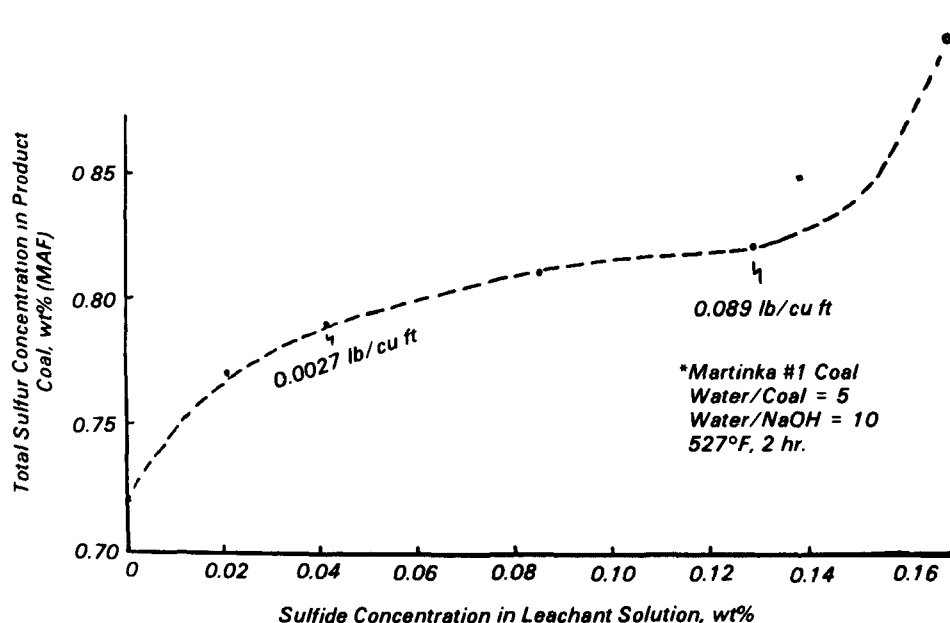


Figure 2. Sulfur concentration in HTT coal versus sulfide concentration in leachant.

### Zinc Oxide

Complete removal of sulfide sulfur from the spent leachant was achieved with zinc oxide, depending on the ZnO/S ratio and temperature. At 176°F and ZnO/S ratios of 3 and 1.75, 100 percent removal was achieved in 10 and 30 minutes, respectively. At a ratio of 1.25, about 85 percent was removed in 60 minutes. At 104°F and ZnO/S ratio of 3, 98 percent was precipitated in 60 minutes.

Potential problems associated with the use of ZnO are: (1) residual zinc in the regenerated leachant, which might contaminate the coal product upon recycle of the leachant, and (2) regeneration of the ZnS for recycle. Total sulfide sulfur removal from the spent leachant for recycle is not necessary. Previous work at Battelle has demonstrated that regenerated leachants containing about 0.12 percent sulfide sulfur can be recycled without any adverse effect on the degree of desulfurization. Therefore,

the problem of residual zinc can probably be minimized by operating at conditions which remove most of the sulfide sulfur without solubilizing an appreciable amount of the zinc.

Because of the cost of ZnO, the ZnS from regeneration of the leachant must be regenerated for recycle. In some other commercially operating processes, zinc is regenerated by roasting under oxidizing conditions. The ZnS reacts with the oxygen to form ZnO and sulfur oxides (SO<sub>x</sub>). The SO<sub>x</sub> are converted to sulfuric acid, and the ZnO is mixed with a reducing agent, generally carbon, and reduced to metallic zinc. Originally, it was contemplated that direct roasting would produce ZnO for recycle. However, after further consideration and discussion with zinc producers, it is considered doubtful that ZnO of the desired particle size can be produced by this approach. Therefore, it may be necessary to roast the ZnS to ZnO, reduce the ZnO to metallic zinc, and then oxidize the zinc metal to ZnO by the American process.

### Iron Compounds

Certain iron compounds are candidates for regenerating the spent leachant. Freshly prepared ferrous hydroxide, Fe(OH)<sub>2</sub>, was the most effective. At a temperature of 77°F and a Fe(OH)<sub>2</sub>/S ratio of 3, 90 - 98 percent of the sulfide sulfur was removed in 60 minutes. Fe(OH)<sub>2</sub> gave a sulfur removal efficiency of about 80 percent under the same conditions. When properly prepared, iron oxide that had been reduced with hydrogen and then partially oxidized (reduced activated Fe<sub>2</sub>O<sub>3</sub>) was effective in removing 80 - 90 percent of the sulfide sulfur. Other iron compounds (e.g., untreated Fe<sub>2</sub>O<sub>3</sub>, Fe<sub>3</sub>O<sub>4</sub>, FeO, metallic iron, and soluble iron compounds) did not remove the sulfide sulfur from the spent leachant.

Regeneration of Fe<sub>2</sub>O<sub>3</sub> from the reacted iron sulfides appears to be technically feasible. Treating leachant with once-generated oxide resulted in the removal of about 85 percent of the sulfide sulfur; twice-regenerated oxide removed about 80 percent of the sulfur. The lower degree of sulfur removal may have resulted from a lower Fe/S ratio—8 as compared to 11.

Freshly precipitated iron carbonate was found to be an effective agent for desulfurizing the spent leachant. At FeCO<sub>3</sub>/S ratios of 1.5 to 10, sulfide sulfur extractions of 80 - 97 percent were obtained with 30-minute treatment times

at room temperature. With an FeCO<sub>3</sub>/S ratio of 1.5, adequate desulfurization for recycle of leachant—80 - 83 percent sulfide removal and 76 percent total sulfur extraction—was obtained.

A method was devised and checked for recycle of the iron values as iron carbonate. This involved: (1) separating the precipitated iron sulfide from the leachant, (2) dissolving the iron sulfide with sulfuric acid solution, and (3) precipitating the iron as carbonate by use of sodium carbonate.

### Other Regeneration Methods

Other leachant regeneration approaches involving electrolysis and the use of activated carbon, soluble iron compounds, lime, and copper were investigated and were found to be unsatisfactory for various reasons.

### Coal Desulfurization

The physical and chemical properties of U.S. coals vary substantially because of differences in rank, mineral composition, maceral composition (organic microstructure), pyritic sulfur content, and organic sulfur content. The desulfurization potential depends on: (1) the content and size distribution of pyrite, and (2) the content and distribution of organic sulfur by functional groups. Pyrite that is finely distributed throughout the coal macerals is difficult to remove by physical means. Organic sulfur that is contained in carbon structures can only be removed by severe chemical treatment. The chemical treatment, necessary to achieve a given residual sulfur value, varies from coal to coal. The optimum conditions for desulfurization can only be determined experimentally.

Near-optimum HT desulfurization conditions were previously established for a number of different U.S. coals by extensive research supported by Battelle. In this project, three coals were selected for testing to these near-optimum conditions: a Northern Appalachian coal (the middling product from the Homer City, PA, coal cleaning plant); a Midwestern coal (an Illinois coal from the Delta Mine); and a Western coal (a subbituminous coal from the Colstrip Mine).

The operating procedure entailed heating an aqueous slurry of the coal and leachant in the miniplant autoclave, and withdrawing samples at intervals. The treated coal was then separated from the spent leachant by a series of washing and filtration steps and vac-

uum dried for analysis. In some cases the washed coal was separated into various size fractions by screening before was dried and analyzed. The test conditions for the desulfurization tests are given in Table 3.

Results of the coal desulfurization tests are shown in Table 4. Treatment of the Northern Appalachian coal with mixed leachant (NaOH/CaO) for 10 minutes at 527°F resulted in the extraction of 94 percent of the pyritic sulfur and 7 percent of the total sulfur. No improvement in sulfur or ash removal was noted by extending the treatment period to 60 minutes.

Treatment of the Midwestern coal with mixed leachants resulted in reductions in the total sulfur content which ranged from 62 to 65 percent. Over the range of conditions tested, increased reaction temperatures and increased reaction times provided only marginal improvements in sulfur reduction. A potentially negative side effect of the hydrothermal treatment was an increase of the coal ash content which results from retention of sodium and calcium from the leachant. However, while these alkali metals increase the ash content, they also have been shown to react with sulfur during combustion to form solid sulfates which are readily collected by boiler particulate control devices.

Treatment of the Western coal only with water produced a lower sulfur ash product than when the coal was treated with mixed leachants or with NaCO<sub>3</sub>. Reduction in the total sulfur and total ash content with water treatment for 10 minutes at 527°F was 30 and 9 percent, respectively.

An evaluation of sulfur content by particle size for tests 95, 93, and 97 shows that (except for the -325 mesh fraction) there is no substantial difference in sulfur level by particle size. This suggests that desulfurization under the process conditions tested is as effective for the 50 × 100 mesh particles as for the 200 × 0 mesh particles.

Both the Northern Appalachian and Western coal data show that a large fraction of the ash is concentrated in the 325 × 0 particle size range. For this condition, the ash content of the product can possibly be reduced by separating out the -325 mesh fraction for further treatment. This treatment might include a weak acid wash which has been shown to be effective in removing residual ash from HTT coal.

### HTT Process Cost Studies

Process construction and operating costs were developed for using the HTT process on an Eastern coal and a Western coal. Cost studies were also performed on a combined physical/HT process using a second Eastern coal. The coals chosen to represent these categories were, respectively, Westland coal, Colstrip coal, and a cleaned middling coal from the Homer City, PA, coal preparation plant.

Some coals (i.e., those containing low concentrations of organic sulfur and high concentrations of pyritic sulfur) can be physically cleaned at low specific gravities (1.3) to produce a low-sulfur coal which will meet the 1971 Federal Sulfur Emissions Standard of 1.2 lb of SO<sub>2</sub>/10<sup>6</sup> Btu for coal-fired steam generators. However, deep cleaning at these low specific gravity conditions is not cost effective because of the small fraction of coal recovered from the process.

To make the process economically feasible, the waste stream from the low specific gravity separation must be cleaned to produce a middling product than can be used in compliance with less stringent SO<sub>2</sub> emission standards. An alternative to this multistream physical cleaning process would be to use a combined physical-hydrothermal process. In the combined process, the sink fraction of the deep physical cleaning process would be chemically cleaned

**Table 3.** Desulfurization Test Conditions

Coal (Test No.)	Particle Size, mesh	H <sub>2</sub> O/coal	NaOH/coal	Reaction Conditions		Temp. °F	Time, Min.
				CaO/coal	Na <sub>2</sub> CO <sub>3</sub> /coal		
N. Appalachian (95)	8 × 0	3.0	0.26	0.20	—	527 <sup>a</sup>	10
Midwestern (86)	50 × 0	3.0	0.26	0.10	—	527 <sup>a</sup>	10, 60
(94)	50 × 0	3.0	0.26	0.10	—	527 <sup>a</sup>	10, 20
Western (91)	50 × 0	3.0	0.03	0.10	—	437 <sup>b</sup>	60
(93)	50 × 0	3.0	0.03	0.10	—	437 <sup>b</sup>	15, 60
(96)	50 × 0	3.0	—	—	0.06	527 <sup>a</sup>	10, 20
(97)	50 × 0	3.0	—	—	—	527 <sup>a</sup>	10

<sup>a</sup>Tests were run at an autoclave pressure of about 900 psig.

<sup>b</sup>Tests were run at an autoclave pressure of about 400 psig.

**Table 4.** Desulfurization Tests Results

Coal (Test No.)	Treatment time, min.	Raw Coal Analysis, wt % <sup>a</sup>				Clean Coal Analysis, wt % <sup>a</sup>				
		Ash (MF)	Total Sulfur	Pyritic Sulfur	Organic Sulfur	Ash (MF)	Total Sulfur	Pyritic Sulfur	Organic Sulfur	
N. Appalachian (95)	10	20.2	2.82	2.6	—	20.4	0.91	0.14	0.64	
	20	20.2	2.82	2.6	0.22	21.9	1.04	0.19	0.68	
Midwest (86)	10	20.5	4.83	1.48	1.70	30.8	1.83	0.16	1.45	
	(86)	60	20.5	4.83	1.48	1.70	33.9	1.64	0.20	1.19
	(94)	10	20.5	4.83	1.48	1.70	33.8	1.71	0.11	1.28
	(94)	20	20.5	4.83	1.48	1.70	33.5	1.68	0.09	1.29
Western (91) <sup>b</sup>	60	10.3	0.92	0.26	0.49	18.0	0.88	0.32	0.51	
	(93)	15	10.3	0.92	0.26	0.49	16.7	0.66	0.18	0.46
	(93)	60	10.3	0.92	0.26	0.49	19.0	0.70	0.26	0.32
	(96)	10	10.3	0.92	0.26	0.49	11.2	0.67	0.17	0.49
	(96)	20	10.3	0.92	0.26	0.49	11.0	0.67	0.17	0.49
	(97)	10	10.3	0.92	0.26	0.49	9.36	0.64	0.16	0.43

<sup>a</sup>Sulfur values on moisture ash free basis. Sulfate sulfur was also determined but are not shown in this table. Organic sulfur is the difference between total sulfur and pyritic and sulfate sulfur.

<sup>b</sup>Average of samples 91-1 and 91-3.

coal to also meet the 1.2 lb SO<sub>2</sub>/10<sup>6</sup> Btu emission limit. The chemically cleaned coal would then be recombined with the deep cleaned (1.3 float product) coal to provide a single compliance fuel.

The basis for the cost estimate was viewed as very critical. Factors such as interest rates, raw material costs, and return on investment can affect the cost estimate markedly. The economic basis of the estimates is explained below.

The HTT process is assumed to be self-contained, generating its own steam, and managing its own operations. The assumption has been made that a mine or utility owns a plant, so that all treatment costs are based on a toll's being placed on the coal processed for processing charges.

Interest costs are assumed to be 10 percent/yr. Plant cost basis was selected as January through March 1978. A debt fraction of 0.6 is assumed for the plant.

The plant's economic life is assumed to be 12 years, construction requiring an additional 3 years. During the three construction years, capital is assumed to be spent in a 20/40/40 percent pattern.

For simplification, during the 15 years of operation and construction, inflation is neglected. The annual income from operations is assumed to be 20 percent before income taxes and interest, and to be constant throughout the economic life of the plant. The capital-related costs, including depreciation, maintenance, property and ad valorem taxes, and inventory taxes, are assumed to be 21 percent of all invested capital.

The working capital in each case is based on the assumptions in Table 5. If working capital were increased to include coal holdings (15-day inventory of product coal), the required capital would have to be increased by \$2.5 million for inventory and \$4.8 million for receivables, less the payables at \$20/ton for coal.

Raw material cost assumptions were generalized from the January-March 1978, *Chemical Market Reporter*. Coal costs are not taken from any single source, but are based on generalized observation of the market.

Capital equipment costs were derived from several sources. When earlier estimates of equipment were considered appropriate, these estimates were included and escalated to the January-March 1978 period. When estimates from other sources were used, these data were escalated to the 1978 basic also. Labor estimates are based on

**Table 5.** Assumptions of Working Capital Requirements

30 day average inventory of raw materials on hand at all times

15 days average product inventory on hand at all times

40 days of receivables outstanding at all times

15 days payables outstanding

No raw coal cost is assessed to product or raw material inventories

(Coal is treated on a toll basis)

$$\text{Working Capital} = \frac{\text{Product Inventory}}{\text{Costs}} + \frac{\text{Raw Materials}}{\text{Inventory}} + \text{Receivables} - \text{Payables}$$

\$16.00/hour, which includes supervision and other related items.

Three case studies, each based on a different coal, were used to derive cost estimates for producing a chemically cleaned low sulfur fuel.

### Westland Coal Processing

The hydrothermal processing of Westland coal using HTT is typical of Eastern coal treatment and is considered to be the case with the least environmental impact with zero liquid discharge. The economics of the process would require a coal cleaning charge of \$37 - \$38/ton of product coal in 1978 dollars. This cost would be added for the coal purchase price. Process costs are expected to continue to increase due to inflation and increasing interest rates. Some process modifications and improvements can be realized by eliminating nickel in the reactor and fire-heater heat exchangers. The ferrous carbonate system could still potentially reduce the cost of processing, although the reduction at this point is not thought to be large unless the FeCO<sub>3</sub> ratio can be reduced. Another alternative, which has not been explored, is to add a sulfuric acid plant instead of a sulfur plant; however, this alternative must be investigated in detail since H<sub>2</sub>S feed purification would be required to make the plant economically viable.

### Colstrip Coal Processing

The Colstrip case is typical of Western subbituminous treatment. Only water and a dispersant for raw chemicals are used to treat the coal rather than a true leachant. Total estimated cost of a ton of clean coal in this case, exclusive of transportation and cost of the coal, is \$10.22.

Some concepts that merit further investigation for this case are: running the solids countercurrent to the liquid in the reactor section as a countercurrent two-stage reactor to increase the untreated water recycle; investigating hydroclones for initial solid/liquid separation to achieve more efficient dewatering; and recovery of metals in the throwaway stream, since rare earth metals, such as vanadium, are present in the coal, and are presumed to leach out of the coal.

### Homer City Coal Treatment (Physical/Hydrothermal)

Economic evaluation of a combined physical/HT treatment plant producing 575 tons/hr of cleaned coal (375 tons by the HT process and 200 tons by physical cleaning) indicated the costs to be about \$24/ton, which includes capital related costs, profit, interest, and taxes, estimated at about \$7/ton.

### Conclusions

By using coarser coals (-20 and -50 mesh, as compared to -200 mesh) and other process modifications, significant process improvements in the liquid/solid separation segment can be realized. Separation rates can be increased by a factor of 10 by adding 0.5 lb of a dispersant/ton of coal processes. Sodium, an undesirable contaminant from the HTT process, can be effectively removed from the coal product with a reasonable number of washing stages using saturated lime water as the washing medium. Washwater consumption is minimized by using countercurrent slurry washing.

The moisture content of the coal product is reduced from about 60 per-



cent to about 40 percent by using centrifugation. Furthermore, the mineral matter content is reduced to below that of the raw coal during the desulfurization step and subsequent downstream processing. The final product is a solid fuel with a reduced mineral matter content and acceptable sodium content. It contains less than the equivalent of 1.2 lb SO<sub>2</sub>/10<sup>6</sup> Btu, and is impregnated with a sulfur scavenger. Previous work indicates that this coal product can be burned with little or no sulfur emission.

Of the leachant regenerates evaluated, zinc oxide, iron and iron hydroxides, reduced activated iron oxide, and iron carbonate removed essentially all or most of the sulfide sulfur from the spent leachant.

Using the process improvements developed under this program, total costs (including capital related costs, profit, interest, and taxes) for a self-contained HTT plant processing 400 tons/hr of coal are estimated to range from \$38/ton of product coal for desulfurization of a typical Eastern coal (e.g., a Pittsburgh seam coal) to \$10/ton of product coal for treatment of a Western subbituminous coal (1978 dollars). These costs include profit, interest, and taxes which are estimated at \$10 and \$4/ton of product for the Eastern and Western coals, respectively.

The estimated cost of the combined physical/HTT treated coal is \$24/ton. Eliminating the middling fraction dryers in the physical cleaning process could reduce this cost. Process modifications recommended in the Westland case would also apply here.

The above costs do not include any credit for sulfur capture by the calcium in the HTT coal during combustion. Previous work for the U.S. EPA (Contract 68-02-2119) indicated that about 50 - 100 percent of the sulfur remaining in the HTT coals was captured during combustion of the calcium impregnated coals in a 1 lb/hr and a 50 lb/hr pulverized coal combustion unit. Thus, it is possible that the HTT process can be used to produce a solid fuel that will meet the revised sulfur emissions standards which require a 70 - 90 percent reduction in SO<sub>2</sub> emissions. However, testing on a larger scale would confirm the sulfur capture that can be obtained in commercially operating boilers.

## Recommendations

Some additional areas of investigation of potential value are:

- Replacing evaporators with reverse osmosis units might reduce water purification costs in the washing section. Membrane resistance at the pH of such solutions have not been investigated. Potential savings are \$2 - \$3/ton of coal produced.
- Precipitation of sulfur from the leachant by a process similar to the citrate flue gas desulfurization process. This would require some chemistry and process studies, but

could save \$5 - \$7/ton of coal in re-generation costs.

- Studies in washing at the same temperature as sulfur leaching. This could reduce the number of washer stages from the currently required 13.

Positive results of such process studies could bring the costs of treating Westland coal, for example, more into line with those normally reported for flue gas desulfurization.

## Conversion Factors

<i>Multiply English Unit</i>	<i>By</i>	<i>To Obtain SI Unit</i>
<i>British thermal unit</i>	<i>0.252</i>	<i>kilogram—calories</i>
<i>British thermal unit/pound</i>	<i>0.555</i>	<i>kilogram—calories/kilogram</i>
<i>cubic foot</i>	<i>28.3</i>	<i>liters</i>
<i>degrees Fahrenheit</i>	<i>0.55(°F - 32)<sup>a</sup></i>	<i>degrees Celsius</i>
<i>mesh number<sup>a,b</sup></i>	<i>-8</i>	<i>2.36 millimeters</i>
	<i>-20</i>	<i>850 micrometers</i>
	<i>-50</i>	<i>300 micrometers</i>
	<i>-60</i>	<i>250 micrometers</i>
	<i>-100</i>	<i>150 micrometers</i>
	<i>-200</i>	<i>75 micrometers</i>
<i>-325</i>	<i>45 micrometers</i>	
<i>pound</i>	<i>0.454</i>	<i>kilograms</i>
<i>pounds/million British thermal units</i>	<i>1.8 × 10<sup>-6</sup></i>	<i>kilograms/kilogram—calories</i>
<i>pounds/square inch (gauge)</i>	<i>(0.06805 psig + 1)<sup>a</sup></i>	<i>atmospheres (absolute)</i>
<i>square foot</i>	<i>0.093</i>	<i>square meters</i>
<i>ton (short)</i>	<i>0.907</i>	<i>metric ton (1000 kilograms)</i>

<sup>a</sup>Actual conversion; not a multiplier.

<sup>b</sup>Mesh numbers correspond to U.S.A. Standard Sieve Series, as specified by ASTM - E - 11 - 70.

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*The complete report, entitled "Process Improvement Studies on the Battelle Hydrothermal Coal Process," (Order No. PB 85-216 588/AS; Cost: \$22.00, subject to change) will be available only from:*

*National Technical Information Service  
5285 Port Royal Road  
Springfield, VA 22161  
Telephone: 703-487-4650*

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