

PRODUCTION OF ASHLESS, LOW-SULFUR  
BOILER FUELS FROM COAL

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SUMMARY

Current and impending restrictions on emission of sulfur dioxide to the atmosphere have brought out the fact that a shortage of low-sulfur fuels exists for power plant and industrial use. This situation has prompted considerable research work on removal of sulfur dioxide during or after combustion (i.e. the so called stack gas treatment processes). Because they have had considerable research emphasis throughout the industry, much of the publicity discussing methods of alleviating sulfur dioxide pollution has been concentrated on stack gas treating processes.

Coal conversion processes have generally been overlooked in this field, since most of them have historically been directed toward conversion of coal to light distillate fuels, and hence have been fairly costly. A much less costly process is now under development, however, which is capable of producing an ash-free, low-sulfur fuel for power plant and industrial use. The process, known as the Pittsburg & Midway Solvent Refined Coal Process, is being developed under the sponsorship of the Office of Coal Research. The process appears to be potentially more attractive than other methods of alleviating sulfur dioxide pollution.

The Solvent Refined Coal Process can produce a low-sulfur, ashless fuel to sell at a considerably lower price than most other low-sulfur fuels. Furthermore, the use of Solvent Refined Coal has a number of potential advantages over stack gas treating processes even at about the same basic overall cost. It appears that the Solvent Refined Coal Process will ultimately play a key role in providing a low-sulfur fuel to meet the requirements of utilities operating under air pollution control laws and at the same time make possible the utilization of current reserves of high-sulfur coal.

INTRODUCTION

The past few years have seen a much greater concern in this country regarding atmospheric pollution, and this has led to increased legislation restricting the discharge of many substances to the atmosphere. Two of the major targets of such legislation have been sulfur dioxide and particulate matter. While restrictions on particulate matter are not new, they are becoming more stringent all the time. Restrictions on sulfur dioxide are more recent, however, and promise to have a much greater impact on the national energy picture. Many of the major cities of the United States now have limitations on the sulfur dioxide content of stack gases and it appears that such limitations will become both more widespread and more stringent. Even in remote areas such as the Black Mesa region of Arizona there is great concern about atmospheric pollution (1). Eventually restrictions on sulfur dioxide emissions will probably be applied to the entire country. On the basis of present trends it appears that these restrictions will limit fuels to a

sulfur content of less than 1% and in many densely populated areas the limits will be less than 0.5%.

Many articles and publications in the past several years have discussed various means of alleviating the air pollution problem resulting from emission of sulfur dioxide to the atmosphere. Most of the research work being done and therefore most of the publications have emphasized only one method of reducing sulfur dioxide emissions; namely, the treatment of stack gases following combustion of high-sulfur fuels. A number of processes have been announced for desulfurization of petroleum residual fuels, but comparatively little has been said publicly about removing sulfur from coal prior to combustion. Even when sulfur removal prior to combustion has been discussed it has generally dealt only with mechanical removal of pyritic materials from the coal. While this can be done in some cases for a moderate reduction of sulfur content, it is not practical for attaining the extent of sulfur removal required for most high-sulfur coals.

When coal conversion processes are discussed in connection with sulfur removal, they are usually dismissed with the contention that they are too expensive. This conception has undoubtedly resulted from the background on the work on coal conversion processes in the past. Research work on direct hydrogenation of coal dates back to the work by Bergius as early as 1913. Most of the early work was directed toward production of gasoline and distillate fuel oils from coal. The early workers very quickly discovered that they could not produce significant quantities of distillate materials by direct hydrogenation without the use of catalysts. Almost from the earliest days, therefore, catalytic processing was used in the direct hydrogenation of coal. For the same reason catalytic processes were also emphasized in the hydrogenation of coal tars produced by the carbonization of coal.

Research work in hydrogenation of both coal and coal tars derived from carbonization of coal has continued intermittently since that time. Even the latest processes developed have not as yet proved to be economically attractive, however, in spite of the improved technology and improved catalysts developed since the early days. All of the research work carried out from 1913 to the present time, primarily using catalytic conversion, undoubtedly forms the basis for conclusions by many observers that sulfur removal by direct conversion is not economical.

#### SULFUR REMOVAL BY COAL CONVERSION

##### 1. Solvent Refined Coal Process

A coal conversion process is currently being developed which is not nearly as costly as earlier processes. The coal is converted not to distillates but to a low-sulfur, ashless fuel resembling pitch in its outward appearance. There are a number of factors which make this process much less expensive than prior coal conversion processes. These factors are (1) a much lower hydrogen consumption (about 1-2% by weight of coal compared to about 6-8% for conversion to distillates); (2) less costly equipment and lower operating costs resulting from the use of less severe operating conditions; and (3) no catalyst is required.

The process is being developed by The Pittsburg & Midway Coal Mining Co. under the sponsorship of the Office of Coal Research, U. S. Department of the Interior. It has been designated as the Pittsburg & Midway Solvent Refined Coal Process.

A schematic flow diagram of the Solvent Refined Coal Process is given in Figure 1. Raw coal is pulverized and mixed with a coal-derived solvent boiling in the general range 550-800°F. The coal-solvent slurry is pumped, together with hydrogen, through a preheater to a reaction zone, or dissolver. The dissolver is operated at a temperature of about 800°F. and a pressure of 1000 psig. At these conditions about 95% of the MAF coal is dissolved. After separation of excess hydrogen, the dissolver effluent consists of the coal solution plus the undissolved inorganic material from the coal. This slurry goes to the filtration section of the plant where the undissolved coal solids are separated. The filtrate is sent to a vacuum flash distillation step for removal of the solvent for recycle. The bottoms fraction from the vacuum flash tower is a hot liquid with a solidification point of about 300°F. This is the major product of the process, and is known as Solvent Refined Coal. This material can either be sent as a hot molten liquid to an adjacent power plant or solidified for shipment to another location.

The Solvent Refined Coal has a heating value of about 16,000 BTU per pound and is surprisingly uniform in its analysis regardless of the type of coal fed to the process. For example, Solvent Refined Coal products derived from both lignite and bituminous coals, when vacuum flashed to the same melting point, have the same carbon-hydrogen ratio, heating value and oxygen content even though the coals from which they were derived differ widely in these characteristics.

There is some variation in sulfur content, however, for Solvent Refined Coal produced from widely different coals. In general all of the pyritic sulfur and over 60% of the organic sulfur is removed during the process. This means that in processing a coal containing 2% pyritic sulfur and 2% organic sulfur (total sulfur content of 4%) the Solvent Refined Coal product would contain considerably less than 1% sulfur. This represents a substantial reduction in potential sulfur dioxide emission. Where the ratio of pyritic sulfur to organic sulfur is higher than 1 to 1, as it is in most cases, the extent of sulfur reduction would be even greater. A total sulfur removal of 85% is not uncommon for the process. Since the heating value of the Solvent Refined Coal is much greater than that of raw coal the effective sulfur content is even lower than it would first appear. For example, Solvent Refined Coal having a sulfur content of 0.8% would be equivalent in sulfur dioxide emission to a Western coal containing 0.4% sulfur and having a heating value of 8000 BTU per pound.

The hydrogen requirement of the Solvent Refined Coal Process is about 1-2% by weight of the coal treated. The hydrogen can be readily obtained by steam reforming of the by-product gas from the process. There is sufficient gas produced to supply all of the hydrogen requirements if process fuel can be obtained from another source. In most cases it is expected that a part of the Solvent Refined Coal produced would be used as process fuel. This would eliminate any requirement for natural gas. In view of the impending shortage

of gas in most areas having large coal deposits, this is a very real advantage.

In addition to the Solvent Refined Coal, a light liquid is also produced in the process. This liquid has a boiling range of about 100-450°F. and is usually about 10-15% by weight of the original coal charged. This product would be suitable as feed stock to a petroleum refinery and should be a very valuable blending component.

The liquid product is considerably higher in nitrogen than petroleum fractions of the same boiling range, but nitrogen removal processes have been and are still being developed in connection with catalytic hydrocracking processes. It is anticipated that the heavier fraction of this product would be processed by catalytic hydrocracking followed by catalytic reforming, while the light liquids would be processed by hydrotreating plus reforming. The high cyclic content of the coal-derived material should provide excellent reformer charge stocks in either case. The reformat would thus have a high concentration of benzene, toluene and xylene. These materials are valuable either as chemical by-products or to provide the aromatic compounds which will be needed in greater quantities for non-leaded gasolines of the future. The light liquid also contains large quantities of phenol and cresylic acids (about 15-20% by weight) which could be separated and sold as by-products. The results of work in other laboratories on similar coal-derived liquids generally support the above observations<sup>(2)</sup>.

The solids from the filtration section are dried to remove excess solvent, then burned in a fluidized combustion zone. The solids contain considerable undissolved carbon (35-55% by weight) and it is desirable to recover this for its heating value. In addition to carbon, the solids also contain about 5-8% sulfur. This makes the sulfur dioxide content of the combustion gases quite high, high enough so that it is feasible to combine this sulfur dioxide with hydrogen sulfide from the solution step in a modified Claus process. The fluidized combustion zone would be a Pope, Evans and Robbins fluidized bed boiler. It has recently been shown that sulfur dioxide from such a boiler can be concentrated in a small part of the total combustion gas. The concentration in this gas stream is over 30 times that obtained in normal combustion<sup>(3)</sup>. Such concentrations should make it even more practical to use the sulfur dioxide in a Claus type reaction. This scheme permits the recovery of essentially all of the sulfur from the process as elemental sulfur.

The steam generated from combustion of the mineral residue plus that from process waste heat can be readily converted to electrical power. The power generated is sufficient not only to supply the coal mine and the processing plant, but would also provide excess power. If the processing plant is located close enough, additional savings could probably be attained by sending the steam directly to the main power plant.

## 2. Other Processing Methods

While the Solvent Refined Coal Process was for some time the only publicly announced process for producing low-sulfur fuels from coal, at least one other coal conversion process for production of low-sulfur fuels has recently been announced<sup>(4)</sup>. This is the H-Coal Process, also sponsored in part by the Office of Coal Research. The major difference between the H-Coal Process

and the Solvent Refined Coal Process is that the H-Coal Process uses a petroleum type desulfurization catalyst in an ebullated bed reaction zone. The ebullated bed is maintained by liquid phase fluidization of the catalyst and this requires a very high rate of internal liquid recycle. Both the catalyst and the high internal recycle increase the cost of the process over the Solvent Refined Coal Process. In general it appears that the Solvent Refined Coal Process will be more economical where the required sulfur levels can be reached without the use of a catalyst. The use of a catalytic process may be desirable when sulfur requirements are in the order of 0.1 to 0.2%, although even in such cases it may still be more efficient to use the Solvent Refined Coal Process as the first stage of a two-stage scheme. The second stage would be a catalytic step in which the feed stock would contain much fewer contaminants than the raw coal.

The Solvent Refined Coal Process has been referred to as "non-catalytic", primarily because a commercial catalyst is not used. There is considerable experimental evidence, however, that the inorganic mineral matter in the coal has a significant catalytic effect in the Solvent Refined Coal Process. While the catalytic effect is certainly not as great as that obtained from a fresh commercial hydrodesulfurization catalyst, the difference is probably not nearly as great after normal deactivation of the commercial catalyst. The extent of the expected difference has not yet been established, but it is this difference which will ultimately determine the feasibility of using a catalytic process for direct hydrogenation of coal.

### 3. New Process Developments

Additional laboratory work has revealed that the use of carbon monoxide and steam to replace part of the hydrogen promises to result in further process improvement. Although this idea had been tried many years ago, it is only recently that advantageous results have clearly been shown for the use of carbon monoxide and steam in processing of coal. This recent work was originated by scientists at the U. S. Bureau of Mines<sup>(5)(6)(7)</sup>. As a result of these promising disclosures, laboratory work was undertaken by Pittsburg & Midway to apply the principle to the Solvent Refined Coal Process. It was found by P&M that the use of carbon monoxide and steam was more effective than hydrogen alone for processing of lignite and subbituminous coals. The results were very promising also for bituminous coal, although the improvements were not as pronounced as with lignites or subbituminous coals. In addition, mixtures of carbon monoxide and hydrogen with steam have been found to be about as effective as carbon monoxide and steam alone.

These results strongly suggest that synthesis gas could be used in place of hydrogen in the Solvent Refined Coal Process. The use of synthesis gas should also have an economic benefit since some of the steps normally used to convert synthesis gas to hydrogen could be eliminated. Another advantage is that the reaction of carbon monoxide and steam in the dissolver forms hydrogen in excess of that used in the solution process, with a consequent enrichment of hydrogen in the recycle gas system. Removal of part of this hydrogen could make it cheaply available as a pure gas for later use in catalytic hydrogenation step. This would not adversely affect the composition of the recycle gas and might, in fact, be beneficial. Other potential advantages are that the coal would not require a drying step and phenolic waste

water could be recycled to provide part of the water for the process. This would also eliminate the necessity for treatment of phenolic waste water.

The potential use of synthesis gas or carbon monoxide and steam in the Solvent Refined Coal Process is a relatively new development and is being further studied at the present time. It is anticipated that the work will ultimately make the process even more economically attractive.

#### 4. Economics of Solvent Refined Coal Process

An economic study has recently been made to determine the price at which the Solvent Refined Coal would have to be sold to attain a 10% discounted cash flow rate of return on investment<sup>(8)</sup>. The price of Solvent Refined Coal is very dependent on credits allowed for by-products of the process. A summary of the effect of by-products on the price of Solvent Refined Coal is given in Table I. If the light liquids, sulfur and electrical power are considered as by-products, the price of Solvent Refined Coal would be about 40¢/MM BTU (Case I). The light liquids are valued at \$3.50 per barrel, sulfur at \$10 per long ton and the excess electrical power at 0.4¢ per KWH.

If the Solvent Refined Coal plant were located in an area where it is impractical to market the light liquid as a by-product, the cost of the Solvent Refined Coal would increase to about 45¢/MM BTU (Case II). In this case the light liquid would be included in the Solvent Refined Coal and would lower its melting point to some extent. If it is also not possible to sell sulfur and electrical power as by-products, the cost of the Solvent Refined Coal would further increase to about 47¢/MM BTU (Case III).

The above prices are based on the assumption that the Solvent Refined Coal plant would be located adjacent to a power plant in the Illinois-Kentucky area. This means that the Solvent Refined Coal would be pumped as a liquid directly to the power plant. If the power plant were located at some distance from the processing plant, about 2¢/MM BTU would have to be added for product solidification, in addition to any transportation costs.

The price range of 40 to 50¢/MM BTU for the Solvent Refined Coal product appears very reasonable in comparison with current prices for competitive low sulfur fuels. For example, the recent price quoted for 1% sulfur No. 6 fuel oil was \$4.10 per barrel (about 65¢/MM BTU in New York Harbor) and \$4.62 per barrel (about 73¢/MM BTU) in the Chicago area<sup>(9)</sup>. Furthermore, even at these prices the quantity of No. 6 fuel oil containing less than 1% sulfur is definitely limited.

Some power plants in the Illinois area have turned to shipping low-sulfur coal from the Western states in an effort to meet the restrictions on sulfur content. While such coal is available at the mine site for a low price, the cost of shipping it for 1000-1500 miles is very high. For example, the total cost of Wyoming low-sulfur coal delivered to the Chicago area is about 55-60¢/MM BTU<sup>(10)</sup>. Although lower negotiated freight rates may bring this down to the range of 45-50¢/MM BTU<sup>(10)</sup>, this is at least as high as the price for Solvent Refined Coal. Because it is virtually free and would not require an electrostatic precipitator, the Solvent Refined Coal is much more advantageous for a power plant even at the same price.

## OTHER METHODS OF SULFUR DIOXIDE CONTROL

### 1. Stack Gas Treating Processes

In most reviews of the general problem of air pollution abatement, stack gas treating processes have been discussed as if they were the only practical methods for control of sulfur dioxide emission. Various groups calling for more research on sulfur dioxide control nearly always specifically mention stack gas treatment processes. Considerable research efforts have already been directed toward removal of sulfur dioxide from combustion gases. These efforts have resulted in the emergence of no less than 21 different stack gas treating processes in various states of development.

Probably the most advanced of such processes are the dry limestone process and Combustion Engineering Corporation's dolomite injection wet scrubbing process. Both of these processes adsorb sulfur dioxide on the dolomite or limestone. No attempt is made to recover the sulfur dioxide and as a result no significant by-products are available for sale.

Another of the more advanced stack gas processes is the Monsanto catalytic oxidation process. This process is much more costly than the other two, but recovers sulfuric acid as a by-product. The value of sulfuric acid as a by-product has diminished somewhat in recent months, however, since the price of sulfur has decreased sharply.

All three of these processes have been installed on a test basis in a few power plants throughout the country. The results of these tests have indicated that actual costs of all of these stack gas treating processes will be far in excess of the generally published cost estimates. The increases are apparently so great that it has been said that costs published even six months ago are now obsolete<sup>(11)</sup>. In addition, the effectiveness of these processes for sulfur removal has generally been less than originally anticipated.

For these reasons no attempt will be made to directly compare stack gas treating costs with the costs of producing a low-sulfur fuel from coal. Information obtained from private sources indicates that the actual cost of stack gas treating processes will be in the same general range as the costs of decreasing the sulfur content of the fuel. It is still far too early to attempt any actual cost comparisons at this time, however, and it appears that experimental work should probably be continued in both types of pollution control methods.

In connection with the relative merits of stack gas treating vs. desulfurization of coal it is of interest to consider certain aspects which would not be directly accounted for in most economic comparisons. In this regard there are certain benefits which would tend to favor the coal desulfurization route if the obvious costs of pollution control were about the same in both cases.

(a) By-products - The potential by-products available from stack gas treating processes are generally limited to sulfur or sulfuric acid. A coal conversion process, however, has the potential for production of many

more by-products than can reasonably be considered in an initial economic evaluation. Ultimate utilization of such by-products through further research and development holds the promise of making the Solvent Refined Coal Process considerably cheaper than stack gas treating processes.

In addition, it seems that the power companies generally prefer not to diversify into the chemical business as they would be forced to do by the installation of any stack gas treating process requiring recovery of by-products such as sulfur or sulfuric acid. At the same ultimate cost it is much more convenient for them to simply buy a low-sulfur fuel.

(b) Uniform Boiler Design - Because of the uniformity of the Solvent Refined Coal regardless of coal source its widespread use for power generation could lead to a uniform off-the-shelf design for power plant boilers. At the present time boilers must be designed for a particular type of coal and when a different type of coal is used, severe operating problems can result. This has happened in the substitution of low-sulfur Western coals in boilers designed for Illinois coals<sup>(12)</sup>. It is almost certain that the use of uniform boilers would lead to cost savings for the power companies, since the cost of producing such boilers should be less than if many different designs were required.

(c) The use of a low-sulfur fuel is a positive guarantee that sulfur dioxide pollution would be controlled at all times. Any down-time in the coal processing plant could be handled by appropriate stock-piling of low-sulfur fuel. With stack gas cleaning processes, however, any interruption in the operability of the process would result in excessive emission of sulfur dioxide to the atmosphere. Thus, the use of a low-sulfur fuel is a more reliable and positive method of pollution control.

(d) Integration with other processes - Even though the economics of a Solvent Refined Coal plant standing alone are attractive, additional economic benefits can be attained by integrating it with other processes in a multi-process complex. For example, the Solvent Refined Coal product can be simply a first stage in an overall process to produce distillate fuels and pipeline gas, as well as a low-sulfur power plant fuel.

An especially attractive combination process scheme is now under study by P&M through a subcontract with Chem Systems, Inc. In this scheme the Solvent Refined Coal is hydrocracked and hydrotreated in two catalytic hydrogenation steps to produce a distillate material suitable for use as feed stock to a petroleum refinery. A coal gasification process is used to produce hydrogen for the three hydrogenation steps in the complex. Methane for pipeline gas is produced in both the gasification and hydrogenation steps. The mineral residue from the Solvent Refined Coal Process is charged to the gasification process to utilize the undissolved carbon.

Preliminary results of this study indicate that all of these processes complement each other effectively so as to make a very promising scheme. A conceptual design and economic study of this scheme is being made by Chem Systems, Inc., the first phase of which is soon to be published by the Office of Coal Research.

## 2. Methane

Probably the ultimate pollution-free fuel is natural gas, and its use by power plants would eliminate sulfur dioxide emissions. This is not a very practical solution to the problem, however, since there simply is not enough gas to supply fuel requirements even for existing power plants, not to mention the large quantity of new power plants which will be required in the future. In fact, it is questionable how long many of the power plants now using natural gas will be able to continue burning this fuel.

A number of processes are now being developed to convert coal to synthetic gas. The costs for producing synthetic gas from coal, however, are almost certain to be higher than the costs of producing a heavier liquid material such as Solvent Refined Coal. These higher costs result from greater requirements in terms of both hydrogen consumption and the process steps necessary for conversion. The conversion of coal to gas will almost certainly become a commercial reality as gasification processes are developed, but it is likely that such gas will be used primarily as a premium fuel where its cost can be more easily justified than in a power plant. Similarly, liquified natural gas imported into this country is so costly that it is very unlikely to be used to a significant extent for power generation. To satisfy the requirements for a low-sulfur power plant fuel, it is much more realistic to use a less costly fuel such as Solvent Refined Coal.

## 3. Nuclear Power

Among the other methods which have been suggested for sulfur dioxide emission control is the use of nuclear power. This is also not a realistic solution since the anticipated demand for electrical power is so great that even with the most optimistic advances in nuclear power generation, fossil fuels will be required for many years into the future. Furthermore, nuclear power has its own peculiar type of environmental problems to be solved.

## CONCLUSIONS

The problem of sulfur dioxide and particulate emissions to the atmosphere can best be solved by removing the ash and sulfur from coal prior to combustion. From a practical standpoint, this can be accomplished by the Pittsburg & Midway Solvent Refined Coal Process, now being developed under the sponsorship of the Office of Coal Research. This process appears to be potentially more attractive than other methods of alleviating the present problem of sulfur dioxide and particulate pollution from power plants. In addition, the Solvent Refined Coal Process can help to relieve the current shortage of low-sulfur fuels and at the same time provide a means of utilizing the high-sulfur coal reserves of the nation.

TABLE I

REQUIRED PRICE FOR P&M SOLVENT REFINED COAL

	<u>Selling Price, Including 10% DCF Return on Investment</u> (Cents per Million BTU)
Case I - Light liquid product sold as refinery feed stock. Sulfur and electrical power sold as by-products.	40.7
Case II - Light liquid combined with Solvent Refined Coal for sale as power plant fuel. Sulfur and electrical power sold as by-products.	45.0
Case III - Light liquid combined with Solvent Refined Coal for sale as power plant fuel. No by-products sold.	47.4

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**FIGURE 1**  
**SOLVENT REFINED COAL PROCESS**

