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**AN AUTOMATED MICROSCOPICAL METHOD FOR THE
CHARACTERIZATION OF PYRITE IN COAL**

Technical Report 16

MASTER

By
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August 1979
Date Published

Work Performed Under Contract No. EX-76-C-01-2030

Coal Research Section
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University Park, Pennsylvania

U. S. DEPARTMENT OF ENERGY

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FE-2030-TR16
Distribution Category UC-90a

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PSU-TR-16

**COAL RESEARCH SECTION
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ABSTRACT

The Rapid Scan system of automated reflectance microscopy, under development at The Pennsylvania State University, is a line scanning device capable of characterizing the pyrite content of a coal. The system combines a sophisticated software base with standard hardware components to provide flexibility in analytical format. As a prepared coal sample is moved automatically beneath the microscope objective, reflectance values from adjacent 2 μm squares are sampled by the photomultiplier tube. These values are stored in computer memory for subsequent calculation of the volumetric proportion of pyrite in the sample. Simultaneously, the Rapid Scan also accumulates a distribution of pyrite chord lengths. In a modification of the Rosiwal linear scanning technique, the number of consecutive readings on a pyrite particle are recorded and stored independently in computer memory.

In order to determine the precision of pyrite volume estimates, a suite of coals with varying pyrite content was analyzed at several sample-point densities. The level of precision achieved was found to be a function of sample density and was always superior to the precision of the conventional, visual point counting technique. From these data, sampling plans to achieve a desired level of analytical precision were designed.

The nature of the pyrite chord length distributions was analyzed with two separate statistical approaches. The first, involving parametric techniques, was ultimately rejected because of the highly skewed nature of the distributions. Alternatively, a nonparametric,

or distribution-free approach was adopted. With this technique the likelihood that two sample distributions were drawn from the same population was determined.

Finally, this information was used in a simulation of crushing operations in the fine-coal cleaning circuit of a preparation plant. A sample of the Upper Freeport seam was crushed to pass an 8 mesh sieve, and then split into its natural size distribution using a series of 21 closely sized sieves. Each fraction was analyzed for its pyrite content and chord-size distribution to determine whether they had changed significantly. Three prominent episodes of size change were observed together with two fractions of anomalously high pyrite content. These indicated the most efficient locations for the gravity separations within the circuit.

The ability of the Rapid Scan system to quickly characterize many samples of a feed coal in this manner, makes it a potentially effective tool in helping to optimize preparation facility operation for the removal of pyrite.

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I. INTRODUCTION

A. An Overview of Automatic Image Analysis Techniques

The volume of literature concerning the automation of image analysis has increased greatly in the past decade (Jesse, 1974a), and excellent bibliographies on the subject have been provided by Cole (1971) and Jesse (1974b; 1976). At present, a large number of systems are available commercially, and doubtless, many more laboratory and research models are in use. Therefore, the intent of this discussion is not to extol the features of a particular system, but rather to compare and contrast important aspects of the three basic types of systems in use today.

Although differences among these instruments are many, they possess a fundamental similarity. Differentiation of the components contained within a sample is always effected by detecting differences in their brightness in either the transmitted or reflected light mode. These variations are then transformed into electrical signals for subsequent computer analysis.

Weibel et al. (1972) and Morton (1972) have separated automated systems into different classes based on the scanning mode employed.

Weibel et al. (1972) used the following tripartite classification:

- A. Class I or 'source-plane' scanners
- B. Class II or 'image-plane' scanners
- C. Class III or 'specimen-plane' scanners

In Class I systems, the scanning movement is made on the sample surface by the light source itself. Some of the earliest attempts to automate conventional microscopical analyses used these 'flying spot' scanners

(Taylor, 1954). They have also been used by Roberts and Young (1952), and by Mansberg (1964), for biomedical research. Belk (1963) and Bloom et al. (1964) used this type of system for the detection of non-metallic inclusions in steel. Otherwise, 'source-plane' scanners have not been widely accepted. This, in part, was due to the limitations imposed by the relatively crude and unreliable instrumentation available at the time as well as other disadvantages inherent in this scanning technique (Weibel et al., 1972).

In Class II systems, the scan is made by a sensor on a projected microscopic image. This 'image-plane' scanning technique includes all television-type systems. These systems have found the most widespread acceptance in research and quality control, primarily due to their diverse analytical capabilities. Examples of their application include materials science (Hougardy and Rose, 1971), nuclear materials testing (Jesse, 1971), medicine and biology (Mertz, 1971; Healey, 1971), and steel analysis (Allmand and Coleman, 1971). An important feature of these systems is an on-line memory which permits an extended lineal analysis for the processing of two dimensional data. These data can then be treated statistically to provide a variety of size, shape, and pattern information. A generalized 'image-plane' scanner is illustrated in Figure 1. The information contained within the microscope image is first converted into electrical signals which are scanned in a voltage-time sequence. Next, the detector units select the information bits belonging to the desired features. Multiple detectors permit the resolution of specified grey bands by setting upper and lower threshold grey values. If it is not possible to separate features solely on the basis of brightness differences,

Figure 1. A GENERALIZED IMAGE-PLANE SCANNER

additional shape or pattern information is needed. The feature can then be compared to various test figures to determine shape parameters (Dinger, 1975). Additional processing and storage is handled by the on-line computer and data can be retrieved according to operator specified formats.

Class III systems are the 'specimen-plane' scanners. Here the light source and sensor remain stationary, aligned along the optic axis of the microscope, while the specimen is moved in a plane perpendicular to that axis. The sensing device employed is usually a photomultiplier tube. These systems are capable of the basic lineal analysis and can also provide certain size information based on intercept or chord length distributions (Underwood, 1968). This type of system has also seen widespread application during the past twenty years and is popular today (McCartney and Hofer, 1955; Bomberger and Deul, 1964; Dodge et al., 1967; McCartney et al., 1971; and, Davis and Vastola, 1976). Figure 2 illustrates a generalized 'specimen-plane' scanning device. The sample is automatically moved beneath the microscope objective as the photomultiplier simultaneously resolves the brightness differences. Information flows directly into an on-line computer where it is stored and processed.

Owing to differences among the types of scans, as well as differences among sensing devices, each system will possess various combinations of the problems inherent in the techniques it employs. The potential user must be aware of these and be able to minimize their effects on the analytical results. Class II, 'image-plane' scanners utilize either Plumbicon or Vidicon tubes for projection of the microscope image. The image is then scanned electronically at a very high density

Figure 2. A GENERALIZED SPECIMEN-PLANE SCANNER

of 'picture points'. As many as 500,000 points may be sampled at a high rate of speed for each field. The actual point-to-point distance is determined by the effective magnification of the instrument. After an entire field of view has been scanned, the sample may be repositioned either manually or automatically.

Several problems exist concerning the proper detection of features in the Class II systems owing to the sensitivity level of the tube itself. Research in recent years has emphasized improvement in these tubes as well as more sophisticated shape and pattern analyses (Hougardy, 1971). In many cases, these additional capabilities are necessary because a disparity as large as an order of magnitude may exist between the size of a feature large enough to be detectable and one large enough for its brightness level to be detected accurately (Cole, 1971). Further, resolution is not constant over the entire tube surface. This variation from the same brightness level, due to position in the field, is known as shading. Other fluctuations in illumination can be caused by both the system electronics and optics. Lenses may represent features located near the optic axis as being brighter than the same feature near the edge of the field. These problems create a transition or 'doubtful' region between two phases which may or may not be detected properly (Cole, 1971). This implies that more accurate results will be obtained when the features of interest are separated widely in brightness levels. Figure 3 illustrates the effect of shading on the proper detection of a feature. Here the boundary threshold has been set for the proper detection of the feature located in the center of the field. The feature on the left appears brighter, and therefore, will be measured larger than its actual size.

- a. threshold too low, feature appears larger
- b. proper threshold setting, feature detected correctly
- c. threshold too high, feature appears smaller

Figure 3. THE EFFECT OF SHADING ON CORRECT FEATURE DETECTION
(Modified from Cole, 1971)

The feature on the right will be measured on a contour which is too dark, and will appear smaller than its actual size.

Another problem which hampers correct detection in 'image-plane' systems concerns the information transfer itself. The amplitude of the video signal will decrease with decreasing size, even if the features are at the same brightness level. This phenomenon is known as the modulation transfer function for video scanners (Houquard, 1971). Figure 4a shows an ideal video signal and a more realistic one (4b) for a scan of two features. Ideally, there would be an instantaneous change of electrical signal from the background grey level to that of the feature. In reality, this change takes place over a small finite time period which represents a distance on the field of view. The result is a 'rounding' or apparent thickness to the feature boundaries. Figure 4 also shows the effect of the threshold on boundary setting on the detection and sizing of the features.

Threshold values (the brightness levels used to discriminate between two components) are determined prior to an analysis, usually by a cursory visual scan of the desired features. The boundaries are set just above and below the brightness range of the feature. If the levels are set improperly, many undesired features will be counted in the analysis. If the threshold is set at level 'a' in Figure 4b, the smaller feature will not be detected at all. Thresholds at 'b' or 'c' will result in the detection of both features, but with an increase in intercept length of the larger one. Thus, the detected size of the feature can vary considerably depending on the threshold level chosen.

Figure 4. IDEAL AND REAL VIDEO SIGNALS AND THE EFFECT OF THRESHOLD SETTING ON THE DETECTION AND SIZING OF A FEATURE
(Modified from Cole, 1971)

Other errors can result from features that extend beyond the fixed measuring area for a field of view. These features cannot be sized properly and contribute spurious measurements to the size distribution data. This problem is known as 'blank frame error' and can be somewhat mitigated by reducing the magnification and accepting the accompanying loss of resolution. Another approach involves the creation of a 'guard area' around the perimeter of the frame. The scanner is capable of probing into this region to measure the actual length of any feature intersected by the measuring frame (Cole, 1971).

Every 'image-plane' system possesses the above detection and sizing errors to a certain extent. Whether or not they have been sufficiently brought under control for a particular application of the system must be carefully determined by the potential user. Despite these drawbacks, 'image-plane' systems provide the only means of obtaining sizes (based on maximum chord, or apparent diameter) and especially the shapes of sample features. They can also provide a visual display of the field being measured.

The Class III or 'specimen-plane' scanners possess several advantages over other systems in regard to the proper detection of features. In this case, only one spot located in the center of each field is measured. The spot remains stationary relative to the light source and the photomultiplier detector. This means that results are virtually independent of the common imaging errors encountered in optics. Field curvature, astigmatism, distortion, and lateral and chromatic aberration all approach zero in the center of the field. Therefore, relatively simple optics can be used and there is no need for the error correction equipment found in other systems

(Weibel et al., 1972). Shading errors are also eliminated. These conditions also permit the diameter of the light beam to be greatly reduced giving the further advantage of minimizing unwanted glare from the specimen surface. This improves both the optical resolution and measurement precision.

The specimen itself is automatically moved in a plane perpendicular to the light beam. This is a relatively slow process when compared to the speed of the electronic scanning beam employed by 'image-plane' systems. Both techniques have their inherent advantages and disadvantages. In the 'image-plane' systems, although each field is scanned with a high density of picture points, only a small portion of the total surface area of the specimen may be covered, depending on the power of magnification used and other factors. Features may appear to be 'clustered' in the sample when in fact they are completely random. This apparent clustering may be due only to inadequate sampling of the specimen. As the number of fields for a complete analysis is chosen before a run is made, that same run may have to be replicated several times in order to determine when an appropriate sampling scheme has been obtained. The clustering effect was reported by Harris et al. (1977) in their study of pyrite occurrences in coal with an 'image-plane' system.

The process of continuously moving the specimen beneath the microscope objective, albeit slower, provides better coverage of the entire specimen surface, and the procedure for arriving at an adequate sampling plan is greatly simplified. The primary disadvantage of this scanning mode is that it limits 'specimen-plane' systems to the basic lineal analysis. Features can only be sized by recording intercept or chord lengths rather than maximum chords (apparent diameters).

Also, these systems cannot provide any shape information. 'Specimen-plane' systems are better suited for the accurate determination of the relative amounts of features, as the photomultiplier can resolve subtle shades of grey that could not be properly detected by other systems with less sensitivity.

To summarize, Class II ('image-plane') and Class III ('specimen-plane') scanners are the two types of systems in use today. The 'image-plane' scanners are generally the more sophisticated, and therefore, the more costly systems. They provide a visual display of the field being imaged and a variety of size and shape information. These systems must cope with a number of important problems concerning the proper detection and sizing of features. As a result, they have become increasingly reliant upon hardware improvements.

'Specimen-plane' systems are generally less reliant on hardware and they avoid most of the imaging problems experienced by the other systems. The scanning is performed by the physical movement of the specimen through the light path of a microscope rather than by an electron beam on a 'picture-tube'. Consequently, these systems can provide only the basic lineal analysis which comprises the intercept sizing of features and their amounts or proportions. In contrast, shape and pattern information is available to the 'image-plane' systems. Moreover, 'specimen-plane' scanners are generally less costly and also less flexible in format.

Numerous examples of each type of system are available commercially and with a wide price range. The perspicacious potential user knows in advance of purchase the exact information required for his particular application. Only then can the most efficient and cost effective system

be chosen. The material presented in this section should provide practical information to aid in comparing the relative strengths and weaknesses of these systems.

B. Applications to Coal Research

In their routine analytical work, coal petrographers determine the proportions of the organic constituents of coal (macerals) and the level of vitrinite (the preponderant coal maceral) reflectivity which is an indicator of rank or degree of metamorphism. This information has been used to establish relations between composition and behavior of coal in research tests and to predict behavior in industrial processes. The work is time consuming and results are always subject to operator variation.

Petrographers also seek information regarding the amount of pyrite and its size distribution in a particular coal. Pyrite is the principal sulfur-bearing mineral in coal and its removal is the goal of all beneficiation processes. If the coal is crushed to finer and finer sizes, more pyrite will be liberated, but the clean coal yield will decrease and the overall cost per ton may rise prohibitively. The presence of pyrite is believed to affect the reaction rate of some conversion processes (Tarrer et al., 1977) and its size distribution plays a role in the production of acid mine drainage (Caruccio, 1970). In these matters, routine pyrite size distribution analyses could provide important information, but they are very tedious as many grains must be carefully measured against the eyepiece reticle of a microscope.

A desire for greater speed and accuracy in the routine analysis work, coupled with the potential for characterizing the pyrite content of coal, stimulated the first interest in automation over 20 years ago.

These early efforts used only 'specimen-plane' scanners for two principal reasons: firstly, the reflected light photometry of a polished coal specimen, which had been popular in Europe for some time, was being accepted in the United States. Second, the relatively advanced technology for 'image-plane' scanning was not developed until the mid-1960's (Cole et al., 1971).

The first automated system for coal analysis was introduced by McCartney and Hofer in 1955. Although the sample was moved automatically, the photomultiplier output was fed through an amplifier to a chart recorder and the subsequent data analysis was performed by manual measurement of the reflectance peak heights. The scan velocity was 0.1 mm per minute and was limited by the slow response time of the chart recorder. A complete 20 mm scan took approximately 3 1/2 hours which was considerably longer than a similar analysis could have been done visually. Various hardware problems were significant factors in this system as many measurements were being made near the limits of performance of the available instruments. Reflectance level distribution curves were plotted for replicate runs on the same coal and for a complete rank suite of coals. The authors suggested a classification scheme based on these curves to aid in predicting the amenability of coals to hydrogenation.

Bomberger and Deul (1964) used a Class III system to characterize the pyrite content of coal sampled from 9 Deister table locations. Although a complete description of this equipment was not published, the electronic counters employed were capable of distinguishing only 8 size classes. Histograms comprising these 8 classes were used in a comparison of pyrite size distributions from two different coals.

The use of these distributions was suggested as a possible replacement for the usual method of calculating the efficiencies of various coal cleaning processes.

In 1968, the AMEDA (Automatic Microscope Electronic Data Accumulator) was introduced (Bayer et al., 1968). It was the first automated system to be used in a steel company research laboratory. This Class III system featured a variable-speed microscope stage drive and an electronic computing system capable of distinguishing 10 size classes. Up to 4 features could be sized during a specimen scan by presetting their appropriate reflectance levels. This system proved its versatility in a variety of uses in the laboratory, ranging from pyrite analysis to the detection of rust spots and pits on various metal samples. The system was capable of sampling 1000 points per second.

In a detailed study, McCartney and Ergun (1969) reported on the system that had been developed at the United States Bureau of Mines. The significant improvement introduced here over their previous system (McCartney and Hofer, 1955), was the storage of all reflectance readings directly on the magnetic tape of a computer. After the analysis, the data could be analyzed using the determined reflectance boundaries of the features of interest. A peculiarity of this system was the specially built microscope stage and motor. Movement was imparted to the sample so that the light beam traced an ever-narrowing spiral path at constant velocity on a circular sample surface. An analysis was completed when the light beam reached the approximate center of the sample. The scan rate was 177 $\mu\text{m}/\text{second}$ and the data acquisition rate was 80 readings/second. The results of the

automated runs were compared to conventional pyrite analyses of the same samples and were found to be within acceptable limits.

Surprisingly, the automated mean pyrite chord length was consistently found to be about one-half that of a visual analysis. The authors believed that this was due to the many small cracks and imperfections present in the pyrite grains. Where the petrographer would ignore these and continue measuring a single chord length, the computer would interpret each small gap as a particle boundary. The authors then introduced a computer algorithm that bridged all gaps up to 10 μm in length. The new size data compared favorably with the visual analysis.

One other difficulty encountered in these experiments was the proper demarcation of particle boundaries. Instead of appearing as sharp reflectance discontinuities, gentle slopes were found in some cases. These were due to the sensing spot landing on a particle boundary and returning a short series of reflectance values that were intermediary to the true values of the two adjacent features. This problem became more serious in a later study (McCartney et al., 1971) when the system was used to calculate the proportions of the organic coal components. When runs were made on -20 mesh and -60 mesh (U.S. Standard) samples of the same coal, it was found that the smaller particle size of the -60 mesh sample contributed too many boundary values to duplicate a conventional analysis within acceptable limits.

Kojima et al. (1974) introduced a 'specimen-plane' system that was completely computer controlled. This system utilized the most sophisticated hardware up to that time. The digital computer was able to distinguish 513 grey levels and a histogram of the entire reflectance

distribution was treated mathematically to calculate the proportions of each maceral group. The stage drive moved in discrete steps and at each sample point the reflectance value was integrated for 10 to 20 milliseconds. This eliminated the inevitable 'smearing' of reflectance values that occurs when the sample is moved continuously. The final value was stored in a reflectance distribution while the stage drive moved to the next sample point. This system also employed a special circuit to integrate the light intensity and reduce the effects of its variation on the reflectance readings. Despite the significant improvements made by this system, the authors felt that a blend of two or more coals still could not be analyzed properly. This was necessary for the authors' particular application of coke strength prediction.

These studies have touched on many potential applications and problems of automated reflectance microscopy. Some of these problems persisted despite the significant hardware and software improvements of the more recent systems. It was the fruition of the versatile minicomputer in the mid-1960's that truly provided the researcher with a new dimension of data acquisition and handling. Consequently, analysis time has been shortened while maintaining a better signal to noise ratio compared to the earlier systems. Unfortunately, the reflectance distribution data still stubbornly defied complete analysis. Different approaches to this problem have been taken by McCartney et al. (1971), Kojima et al. (1974), Davis and Vastola (1976), and Hoover et al. (1976). Yet more study is needed in order to understand the relationships among the maceral groups so the reflectance

profile or reflectogram can be effectively deconvoluted. Particle boundaries affect both the maceral and pyrite analyses and also need more basic research.

As every system has its unique features, it is the responsibility of the researcher to be certain that the exact conditions for data acquisition and its subsequent manipulation always accompany the results. Only in this way can the data generated by many different systems be evaluated critically.

II. THE RAPID SCAN SYSTEM OF AUTOMATED REFLECTANCE MICROSCOPY

A. Design, Cost, and User Considerations

In recent years, the desire for a greater sophistication of automated analyses has engendered substantial hardware and software improvements. Many variations of the basic system components that were shown in Figures 1 and 2 can be obtained commercially and most systems can be modified to meet user specifications by the addition of various modules and analytical packages. In general, the degree of sophistication required is reflected in the cost of the system. 'Image-plane' scanners are dependent upon the addition of new hardware modules to mitigate the ever present detection and sizing problems. Consequently, these systems can become obsolete and the user must be prepared to purchase the latest innovations. Another implication of hardware sophistication is that many fine calibrations must be made prior to every analysis. Hougardy (1974) reported analytical errors of up to 100% when these calibrations were made by inexperienced operators. The Rapid Scan, a 'specimen-plane' scanner being developed at The Pennsylvania State University, addresses all of these problems, namely, high cost, hardware dependence, and complexity of routine operation.

The solution to these problems lies in the development of sophisticated software packages to control scanning, data acquisition, and data processing. In this way, flexibility can be built into a class of systems that had been previously limited by a rigid analytical format. It also means that the Rapid Scan cannot become obsolete as the software can be easily modified and updated. Another advantage to software control is that the system is compatible with general

purpose computers. Most 'image-plane' scanners require specially built, completely dedicated computer equipment and other specialized hardware. The major hardware components of the Rapid Scan system are of standard design and can be purchased from a number of manufacturers. Both the software emphasis, and the use of readily available hardware components help to keep the purchase cost low compared to most 'image-plane' scanners.

Because of the software control, little specialized knowledge is required for routine operation. The operator must calibrate the system with the standard of known reflectance value, align and focus the sample, and enter the appropriate run parameters at the videoterminal. There are no other precise calibrations to be made in the course of a data run.

The hardware and software evolution of this system has been reported by Davis (1975), Davis and Vastola (1976), and Hoover et al. (1976). Its analytical versatility also has been demonstrated in these papers. Davis (1975) showed how differences in the pyrite size distributions of two coals could be detected by the automated analysis. The early reflectance distributions of the organic coal components also showed changes when coals of differing maceral composition were analyzed automatically. These analyses were subsequently expanded and improved. Davis and Vastola (1976) compared the visually obtained pyrite chord size distributions of 5 selected coals with those done automatically. Both sets of data showed relatively larger pyrite sizes in two of the coals.

Davis and Vastola (1976) and Hoover et al. (1976) also demonstrated the utility of this system for the measurement of vitrinite

reflectivity as an indicator of rank. The Rapid Scan records only random vitrinite reflectance values as opposed to the maximum values preferred by petrographers when the analysis is performed conventionally. This is due to randomness of particle orientations and the fact that the microscope stage is not rotated to get a maximum value (vitrinite is anisotropic in most coals) during an automated analysis. The random reflectance values can be corrected by a simple algorithm to give the proper maximum values. Davis and Vastola (1976) reported the change in location of the median vitrinite reflectance value as a function of coal rank as well as the changing response of the reflectograms as a whole to different maceral compositions. Hoover et al. (1976) introduced techniques of curve matching to these distributions in order to extract the volumetric contribution of each maceral group.

Since that time, improvements to the system have allowed further data refinements and more analytical flexibility. The ultimate goal is to develop a system that will provide meaningful data, at a reasonable cost, for research, routine analytical work, and quality control applications. This report will explain the present capabilities of the Rapid Scan system in the mode of automated pyrite analysis.

B. System Plan and Explanation

A complete schematic diagram of the Rapid Scan system of automated reflectance microscopy appears in Figure 5. The system comprises 4 principal components:

- A) the microscope photometer
- B) the low-pass amplifier/filter
- C) the computer
- D) the automatic stage drive.

Figure 5. THE RAPID SCAN SYSTEM OF AUTOMATED REFLECTANCE MICROSCOPY

The optical equipment employed is a Leitz Orthoplan MPV-2 microscope photometer with a 100-watt, quartz halogen white light source. The portion of incident light that reflects from the object surface passes a 546 nm filter before impinging upon the photomultiplier cathode (EMI Electronics S-20, model 9659B). The adjustable photomultiplier aperture is set in the shape of a square with an effective measurement area of $4 \mu\text{m}^2$ at a magnification of 625x. The photomultiplier output in milliamperes is converted by a Leitz amplifier system into voltage (0 to +10v) form. The signal then enters the low-pass filter.

A schematic diagram of this specially built low-pass filter can be found in Hartmann (1976, p. 29). This unit is designed to improve the signal to noise ratio by reducing the effective bandwidth, and therefore, blocking any spurious high frequency signals. The maximum voltage output of this device is limited by a 6 volt Zener diode. This makes the output more compatible with the analog to digital converter of the computer which operates in the range of 0 to +5 volts. Incoming signals with a value between +5 and +6 volts are all assigned the maximum digital value by the computer.

The computer facility is a Digital Equipment Corporation model PDP8/A with 32K (thousand), 12-bit words of random access memory. The machine also has the FPP-12 floating point processor which greatly increases the speed of data handling. The 10-bit AD8-A analog to digital converter initially processes all incoming signals by assigning them appropriate values between 1 and 1024. The converter has 16 channels of input available, although only 2 are used in a normal data run.

Several peripheral devices provide flexibility in data output format. The Digital Equipment Corporation VT-52 videoterminal and LA-36 Decwriter are the main interactive units. All data may be quickly displayed on the videoterminal and a high quality hard copy may be obtained from a Xerox 1700 printer. Hard copy graphics are provided by a Houston Instruments DP-1 incremental plotter.

Data storage is accomplished in two ways. Most of the data files are permanently stored on RK05J removable cartridge disks. Each cartridge has a storage capacity of approximately 1.6 million 12-bit words of on-line data. The computer also has the RX8 dual floppy disk system which serves primarily as a back-up. Each of these disks has a storage capacity of 128,000 12-bit words of on-line data.

Another specially designed hardware component in the system is the KIM-1 microprocessor which employs a MOS Technology 6502 microprocessor chip. This device permits two-way communications with printers and videoterminals on 4, 20-milliampere current loop circuits. Five Transistor-Transistor Logic (TTL) level ports are also available for interfacing with other microprocessors. The microprocessor provides a great deal more flexibility, at less cost, than the commercially available computer accessories.

In an automated pyrite analysis, the microscope stage is powered by a Femco AMEDA stage drive, which was first reported by Bayer et al in 1968. The stage is moved continuously (as opposed to discrete steps) and its horizontal and vertical movements are automatically controlled. The drive unit is connected to the microscope on the coaxial operating knobs of the microscope mechanical stage.

The six available scanning speeds range from 25 $\mu\text{m}/\text{second}$ to 800 $\mu\text{m}/\text{second}$. The horizontal scanning distance may be varied from 5 mm to 75 mm. The traversing distance between each scan line may be varied from 8 μm to 1200 μm , depending on the density of sample lines desired.

All of the major hardware components used in the automated pyrite analysis are illustrated in plate 1.

RAPID SCAN HARDWARE FOR AUTOMATED PYRITE ANALYSIS

PLATE I

III. ANALYTICAL CAPABILITIES OF THE RAPID SCAN SYSTEM

A. Equipment Calibration and Sampling Plan

Just as in conventional microscope photometry, the Rapid Scan system must be calibrated with glass standards of known reflectance value. The computerized standardization routine samples the analog to digital converter output, averages many values, and returns a mean reflectance value to the videoterminal. This value may be adjusted with the potentiometer that is built into the low-pass filter component. In this way, the maximum of +5 volts allowable by the computer can be made to correspond to any full-scale reflectance value. A simplified flowchart of the standardization routine appears in Figure 6.

Once the sample is properly aligned, the analysis can begin. Figure 7 indicates the sampling plan used on the usual pelletized, polished crushed coal specimen (ASTM D2797-72, 1978). The horizontal scanning distance (X) is adjusted to inscribe a rectangular grid on the sample surface. The traverse-to-traverse distance (Y) is variable and depends on the number of sample points desired. After the stage drive is started manually, the computer takes immediate control of the data acquisition.

Upon completion of a horizontal traverse, a 9 volt signal is sent on another input channel of the analog to digital converter. The computer switches channels and begins to monitor this signal. Termination of the signal indicates that the traverse-to-traverse movement has been completed and cues the computer to begin sampling a new traverse on the main data channel. After the specified number of readings has been taken on this traverse, channels are switched

Figure 6. A SIMPLIFIED FLOWCHART FOR CALIBRATING THE RAPID SCAN SYSTEM WITH A GLASS STANDARD OF KNOWN REFLECTANCE VALUE

- a. Beginning of Data Acquisition on Traverse
- b. End of Data Acquisition, Check for End of Scan Signal
- X. Total Horizontal Scanning Distance (Variable)
- Y. Traverse to Traverse Distance (Variable)

Figure 7. SAMPLING PLAN FOR PREPARED COAL SPECIMEN

again, in anticipation of the end of scan signal. No data readings are recorded until the traverse-to-traverse movement has been completed.

At the beginning of each new traverse, the computer is delayed about .5 second to allow the stage drive to reach a constant velocity before the data acquisition begins. The actual data acquisition portion of each traverse lies between points 'a' and 'b' marked in Figure 7.

This sampling plan has been found to be superior to one used in earlier versions of the Rapid Scan system. In the older plan, output channels were switched after every reading to check for the end of scan signal. If the signal was not present, another reflectance reading was processed on the main data acquisition channel. This constant switching was found to be too 'noisy' and resulted in incorrect digital values for the reflectance readings.

The new sampling plan allows an uninterrupted series of readings to be taken before the channel switching takes place. The improved reliability of the analog to digital converter in the PDP-8 (over an older PDP-9 computer used previously), combined with more efficient ASSEMBLER level acquisition routines and the improved sampling plan have effectively eliminated this source of error from the analysis.

B. Determination of the Volumetric Contribution of Pyrite in a Pelletized Coal Sample

The automated pyrite analysis is based upon the ability of the system to distinguish among epoxy binder, coal, and pyrite solely on the basis of their reflective properties. The epoxy binder used in the routine preparation of polished coal samples normally comprises 20% to 30% of the sample by volume and it reflects less than 0.1% of

incident light. The first reflectance peak (A) in Figure 8 represents the distribution of the binder in a sample of PSOC-634, the Upper Freeport seam. This was a specially prepared pellet containing greater than 50% of the binder by volume. As the amount of binder varies from sample to sample, its contribution to the overall reflectance distribution must be removed subsequent to the completion of the data acquisition. This is accomplished by assuming a normal (Gaussian) distribution of binder reflectance values and matching a best-fit curve to each distribution. The population of this curve is subtracted from the total, leaving coal, pyrite, and other mineral components in the distribution. The area marked 'B' in Figure 8 represents the total contribution of all organic and inorganic coal components in this sample, other than pyrite. The liptinite maceral group has the least reflectance power and may reflect as little as 0.1% of incident light in oil as determined by conventional means. The inertinite maceral group comprises the most highly reflecting organic coal components and these rarely exceed 6% reflectance in oil. The prominent normal distribution (C) appearing in the coal range in Figure 8 is the contribution of vitrinite, the principal coal component. All of the coal mineral components, exclusive of pyrite, are superimposed on the coal range and their contribution to an analysis varies greatly. Unfortunately it is not possible to subtract their reflectance distributions from the total so that all data are reported on a mineral matter contained basis.

Ramdohr (1969, p. 782) determined that the reflectivity of pyrite in oil was 46% when an unspecified green wavelength filter was inserted in the light train. In polished coal samples, the reflectivity of

Figure 8. THE REFLECTANCE DISTRIBUTION FOR PSOC-634, THE UPPER FREEPORT SEAM,
AT 8% FULL-SCALE REFLECTANCE

pyrite seldom reaches this level as the presence of irregular surfaces, inclusions, and discontinuities in a grain reduce its reflectance power. Regardless, the proper automatic discrimination of pyrite grains from coal particles is easily effected.

The full-scale reflectance value for an automated pyrite analysis is set at 8% for the +5 volt maximum value. This 8% limit acts as the reflectance threshold boundary between coal and pyrite. Invariably, the pyrite grains reflect greater than 8% of incident light, so their voltage outputs are always limited by the Zener diode in the low-pass filter component (see Section IIB and Figure 5). When their signals are digitized, the readings on pyrite are assigned the maximum value of 1024 (2^{10}). Thus, all pyrite readings are stored in the last array element of the reflectance distribution. These are illustrated by 'D' in Figure 8. Examination of the entire reflectance distribution for this coal shows that the greatest reflectance value, other than pyrite, is approximately 3%, and that no readings lie between the 3% value and the 8% storage area for pyrite.

It is very advantageous to set the pyrite boundary as low as possible (8%) because this allows the maximum precision in the 1024 element array. Each category then represents $8/1024$ or about .008% of reflectance difference. Because the epoxy binder reflects less than 0.1%, approximately the first 12 categories hold this distribution and make possible the proper distinction between the binder and the low-reflecting liptinite macerals. If the pyrite threshold had to be established near its theoretical reflectance power (46%), each array element would then represent $46/1024$ or .045% of reflectance

difference, and proper distinction between binder and coal would be very difficult.

Using these relationships, the volumetric contribution of pyrite in a sample is easily calculated. For the distribution in Figure 8, a sample size of 50,000 points was used. After subtracting the best-fit curve estimate of the epoxy binder contribution, 20,185 readings remain in the array. The last array element holds 963 pyrite readings so that its contribution is $963/20,185$ or 0.48% pyrite by volume in this sample.

C. Determination of Pyrite Size Distribution by Lineal Analysis

The theoretical justification for a lineal modal analysis was first demonstrated by Rosiwal in 1898. A random traverse line placed on a randomly selected plane of polish, may also be regarded as being randomly placed within the three dimensional structure itself. Therefore, the lineal fractions obtained provide direct estimates of volume fractions (Hilliard, 1968). Rosiwal measured the intercepted length of each feature and calculated the total distance traversed within each at the end of the analysis. Dividing this length by the total length traversed determined the lineal and, thus, volumetric contribution of each phase.

A simplified lineal analysis for a 3-phase system is shown in Figure 9. As the traverse proceeds across the field from left to right, every intercept is measured and recorded. In the figure, an intercept length of 23 mm is recorded in phase A. Next, a length of 5 mm is recorded in phase B, and so on, until the traverse is completed. The subtotals (L_A , L_B and L_C) are then computed as well as the total length of traverse (L_T). The volumetric contribution of

INTERCEPT LENGTHS

L_A	L_B	L_C
23mm	5mm	25mm
<u>13mm</u>	<u>12mm</u>	<u>-</u>
36mm	17mm	25mm

$$L_T = L_A + L_B + L_C = 78\text{mm} = \text{Total Line Length}$$

VOLUMETRIC CONTRIBUTIONS

$$V_A = L_A / L_T = 36\text{mm} / 78\text{mm} = 46.2\%$$

$$V_B = L_B / L_T = 17\text{mm} / 78\text{mm} = 21.8\%$$

$$V_C = L_C / L_T = 25\text{mm} / 78\text{mm} = 32.1\%$$

Figure 9. A SIMPLIFIED LINEAL ANALYSIS OF A 3-PHASE SYSTEM

phase A can then be computed with the formula $V_A = L_A/L_T$, where L_A is the total length recorded in phase A and L_T is the total length of traverse. This calculation is repeated for the remaining phases in the sample.

There were three important parameters established by Rosiwal as the basis of this analysis:

- 1) the length of the measured traverse must be at least 100 times the length of the coarsest grain;
- 2) the number of traverses necessary is a function of the structural characteristics of the specimen; and
- 3) two traverses should not intersect the same grain.

Interestingly, Rosiwal did not envision the traverses as comprising a parallel set, but as randomly placed lines on the specimen surface. It remained for Chayes (1956) to show that parallel placement of lines was also a necessary condition for the analysis. These four conditions have been fulfilled by the sampling plan that was illustrated in Figure 7.

Initially, the lineal analysis gained little acceptance. This was due primarily to the fact that the method was very time consuming as each intercept length had to be carefully measured with an eye-piece reticle and manually recorded. The development of devices that automatically integrated the distance traversed within each constituent greatly reduced the time factor, and hence, popularized the technique. Such instruments were introduced by Shand (1916), Hunt (1924) and Heinrich (1956, pp 9 and 10). The most popular of these was the Integration Stage of E. Leitz, Inc. (see Galehouse, 1971, pp. 388-389). This microscope stage could be driven by any

of six micrometer spindles. The spindle that corresponded to a particular component was turned whenever that component appeared under the ocular cross-hair, automatically recording the appropriate length.

Despite these improvements, the lineal analysis was almost completely supplanted by point counting (Glagolev, 1934; Chayes, 1956) in the mid-1950's. Today, point counting is the most widely used technique because it requires the least amount of effort to attain a specified sampling error (it is the most efficient technique) (Underwood, 1970). Point counting is also less time consuming than other techniques.

Renewed interest in the lineal analysis is an indirect out growth of the semi-automatic recording devices and the volumetric pyrite analysis introduced in the previous section is simply the total automation of the Rosiwal technique. The high speed and versatility of the computer allows a greater total traverse (L_T) to be measured than is practical by any manual or semi-automatic means. A higher sample density (more sample lines) per unit time means that more of the sample surface is covered and that the precision of the analytical results is increased. Thus, time and precision are no longer the limiting factors of this technique.

A further modification of the original Rosiwal analysis provides another advantage to the automated method. The computer is capable of accumulating a pyrite chord size distribution while the volumetric analysis is in progress. This is achieved by storing, in memory, the length of every discrete pyrite chord immediately after it has been measured.

A practical problem arises in pyrite chord measurement concerning the definition of a discrete particle, or uninterrupted chord length. This was first reported by McCartney and Ergun (1969) when they found that their automated results were not comparable to those derived by conventional means owing to cracks and discontinuities within pyrite grains. This prompted the development of an algorithm that would automatically bridge 'gaps' of up to 10 μm . This same problem applies to small discontinuities occurring between grains as well as those within them. Pyrite occurs in a variety of forms (Reyes-Navarro and Davis, 1976), and when these are studied microscopically, the meaning of the term 'particle' becomes vague. The practical definition adopted for the present study requires that discontinuities either within or between grains must exceed 20 μm before a final chord size measurement is stored in memory. One feature of the computer software is that this value is an operator input variable. This means that any value ranging from 2 μm to approximately 1 mm may easily be substituted for 20 μm at the discretion of the operator.

The chord measurement technique is illustrated in flowchart form in Figure 10. First, a reflectance value is registered. Then, the computer checks to see if the value indicates whether or not a pyrite particle is being scanned. If so, the value is stored temporarily, a flag is set, and another reading is taken. This is illustrated by the right hand branch in Figure 10. The pyrite chord length is measured by summing the number of consecutive readings on pyrite before a 'gap' of greater than 20 μm is encountered. By

Figure 10. A SIMPLIFIED FLOWCHART FOR THE COMPUTERIZED MEASUREMENT OF PYRITE CHORD LENGTHS

definition, gaps whose length is less than 20 μm (10 readings) are included in the chord measurement.

If the value read from the analog to digital converter indicates that something other than pyrite is being scanned, the algorithm becomes more complicated. First, the condition of the pyrite flag is checked. If the flag is down, we are truly on a constituent other than pyrite. In this case, another reading is taken. This is the left-hand branch of Figure 10. If, however, the pyrite flag is up, this indicates that a 'gap' in the pyrite grain is being crossed. Next, the length of this gap is checked; if it is less than 20 μm , another reading is taken. If it is now greater than 20 μm , a chord length measurement has been completed and this value is stored. The pyrite flag is set 'down', the counters are reset, and another reading is taken. These processes are shown in the main trunk of the flowchart in Figure 10. From this figure, it can be seen that for any given analog to digital converter value, one of four major processing branches may be taken. Each branch differs in the number and kinds of statements to be executed. Each branch, therefore, has a different real time of execution. These differences must be equalized by the insertion of several dummy steps along all but the longest branch of the algorithm. Since the stage drive moves continuously at a constant velocity, each branch must take the same amount of time so that each photomultiplier reading is exactly adjacent to the previous one. If a reading is taken too soon, the sample squares will overlap, if too much time elapses between readings, a space will occur between each. Both of these conditions would result in an

incorrect length measurement. The correct spatial relationship that must be achieved between consecutive readings is illustrated in Figure 11.

The fact that the stage drive moves continuously thus creates programming problems relating to the timing of consecutive readings. This is definitely a disadvantage of the present system, but it could be mitigated by certain hardware improvements, such as a computer-controlled stage drive. The principal FORTRAN data acquisition program for the automated pyrite analysis is given in Appendix A.

The following 3 illustrations, Figures 12-14, depict the actual computer interpretation as the photomultiplier scans across different pyrite morphologies. A photomicrograph of the scanned surface appears above each computer interpretation to help the reader visualize how one is derived from the other. For these demonstration scans, the coal/pyrite threshold was set at 2% reflectance so that the constituents with very low reflectance power would appear on the graph. The direction of scan is from left to right and the exact line of scan is indicated on each photomicrograph. The abscissa of each graph shows the actual distance in microns. The ordinates show reflectance levels and the corresponding digital values from the analog to digital converter. The photomicrographs were taken at a magnification of 625X using the optical equipment described in Section IIB.

The pyrite occurrence in Figure 12 can be described as a partially fused framboidal mass. In this case, three pyrite chord lengths were measured by the computer, namely a 2 μm chord, a 22 μm chord, and a 20 μm chord. These are defined by the three prominent

Consecutive Readings for Chord Length Measurement

Figure 11. THE CORRECT SPATIAL RELATIONSHIP BETWEEN CONSECUTIVE PHOTOMULTIPLIER READINGS FOR INTERCEPT MEASUREMENT

Figure 12. COMPUTER INTERPRETATION OF PARTIALLY FUSED FRAMBOIDAL PYRITE MASS

Figure 13. COMPUTER INTERPRETATION OF A PYRITE BLEB

Figure 14. COMPUTER INTERPRETATION OF PYRITE EUHEDRA OCCURRING AS CELL FILLINGS

peaks that reach the coal/pyrite threshold level in the figure. Assuming the length of discontinuity to be bridged was 20 μm , only two pyrite occurrences were stored in the chord size distribution. Because the 2 μm chord and the 22 μm chord are separated by only 4 μm , a pyrite occurrence of 28 μm was recorded. The distance between this chord and the remaining one is about 26 μm , which is greater than the 20 μm limit. This means that the length of the remaining chord, 20 μm , was stored separately.

In Figure 13, a single, irregular pyrite bleb has been scanned. The scanning line has crossed an uninterrupted 30 μm chord. There were no other pyrites within 20 μm on either side of this occurrence, so that a single 30 μm chord was stored. This length is indicated by the prominent peak on the graph in Figure 13. The spike in the graph that narrowly misses the pyrite threshold, represents the coal maceral fusinite. This is the relatively bright (reflecting about 1.8% of incident light) component just to the left of the pyrite bleb in the photomicrograph.

The third pyrite occurrence, shown in Figure 14, is a mixture of euhedral pyrite crystals and irregular blebs filling the cell cavities of fusinite. The line of scan has crossed 4 euhedra. The first two, 6 μm and 14 μm each, are separated by only 6 μm and this occurrence is recorded as a 26 μm chord. The two occurrences on the right, 12 μm and 6 μm each, are separated by about 14 μm and this occurrence was recorded as a 32 μm chord.

One can readily see the effect that bridging discontinuities has on the actual chord measurement. For a given study, this bridging

length should remain constant. The value can be easily changed to accommodate a different method of sample preparation or the particular requirement of a research project.

Samples of the computer-generated data sheets from the automated pyrite analysis appear in Appendix B. The two-page format provides a summary of the information presented in this chapter for each individual analysis. The first page records the exact operating conditions for the analysis, so that they can be exactly reproduced for any number of desired replications. This page also shows the information concerning the amount of pyrite in the sample including the number of pyrite particles encountered, and the total pyrite chord length. The remainder of the page illustrates ways of examining the chord-size distribution, including a logarithmic transformation of the data (see Section VA). Page 2 of the print-out reproduces the pyrite chord-lengths encountered during the analysis of the form of a histogram. Thus, all the data are made readily available for any further qualitative or quantitative analyses desired by the researcher.

IV. ANALYSIS OF DETERMINATIONS OF THE PYRITE VOLUMETRIC CONTRIBUTION IN SELECTED COAL SAMPLES

A. Precision of Volume Fraction Estimates

A suite of five coals, whose pyrite content ranged from 0.34% to 2.33% by volume, was selected for this study. Each analysis was replicated five times using a point density of 100,000 (12 traverses). The analysis was then repeated at point densities of 200,000 (24 traverses), 400,000 (50 traverses) and 800,000 (100 traverses), for a total of 100 sample runs. The data analysis was performed in light of two objectives:

- 1) determination of the reproducibility of results (precision) by replication, as a factor of the number of sample points.
- 2) determination of an efficient sampling plan to achieve a specific accuracy.

A data summary for these analyses appears in Table 1.

In Figure 15, the sample standard deviation (s) for each group of 5 runs is plotted against the total number of sample points. The standard deviation was calculated according to the following formula:

$$s = \frac{\sum_{i=1}^n (\bar{x} - x_i)^2}{n-1}$$

where x_i = an individual observation

$\bar{x}$ = mean of n observations

n = total number of observations

Table 1. Results of Replicated Pyrite Analyses at Selected Point Densities for 5 Coals.

SAMPLE NO. (PSOC)	SAMPLE POINTS	VOL. % PYRITE	($\bar{x}$) PYRITE %	STANDARD DEVIATION (s)
724	100,000	.97 .27 .22 .68 .17	.46	.35
724	200,000	.45 .48 .40 .77 .34	.49	.17
724	400,000	.40 .60 .52 .45 .47	.49	.08
724	800,000	.46 .42 .37 .55 .48	.46	.07
773	100,000	.45 .36 .47 .67 .46	.48	.11
773	200,000	.39 .45 .45 .52 .42	.45	.05
773	400,000	.55 .46 .48 .51 .46	.49	.04

Table 1. Results of Replicated Pyrite Analyses at Selected Point Densities for 5 Coals (Continued)

SAMPLE NO. (PSOC)	SAMPLE POINTS	VOL. % PYRITE	($\bar{x}$) PYRITE %	STANDARD DEVIATION (s)
773	800,000	.43 .39 .41 .40 .42	.41	.02
678	100,000	2.78 2.64 1.86 2.29 1.85	2.28	.43
678	200,000	2.38 2.56 2.25 2.35 2.21	2.35	.14
678	400,000	2.19 2.32 2.12 2.35 2.36	2.27	.11
678	800,000	2.42 2.37 2.32 2.15 2.38	2.33	.11
767	100,000	.47 .21 .40 .42 .57	.41	.13
767	200,000	.41 .36 .37 .35 .48	.39	.05

Table 1. Results of Replicated Pyrite Analyses at Selected Point
Densities for 5 Coals (Continued)

SAMPLE NO. (PSOC)	SAMPLE POINTS	VOL. % PYRITE	($\bar{x}$) PYRITE %	STANDARD DEVIATION (s)
767	400,000	.36 .34 .34 .45 .32	.36	.05
767	800,000	.30 .31 .30 .40 .40	.34	.05
651	100,000	.69 1.22 .89 .77 .68	.85	.22
651	200,000	.91 .71 .76 .69 1.15	.84	.19
651	400,000	.97 .92 1.03 .77 .82	.90	.11
651	800,000	.89 .94 .98 .90 .95	.93	.04

Figure 15. STANDARD DEVIATION (s) OF PYRITE CONTENT VS LOG NUMBER OF SAMPLE POINTS FOR 5 SELECTED COALS

According to the formula, as the number of observations was increased, the sample standard deviation should have decreased as a square root function. In Figure 15, coals 3, 4 and 5 most closely approximated this function. Coal 2 shows a fairly constant decrease in standard deviation indicating that further precision would be gained by increasing the number of sample points beyond 800,000. In the case of coals 3, 4 and 5, the curves show an inflection point at 200,000 sample points, signifying that very little is to be gained by subsequent doubling and quadrupling of point density. In the final case, coal number 1 shows an anomalously high standard deviation at 400,000 points. This value is actually higher than the one for 200,000 points. As each value on the graph represents 5 replicated analyses, this value could not be easily dismissed. When taken in conjunction with the overall irregular shapes of curves 1 and 2, it was concluded that factors beside sample point density were affecting the precision of analytical results. The major sources of this variation were subsequently identified and examined.

In Table 2, the volumetric contribution of the epoxy resin binder for each of the five coals is shown. As the binder routinely comprises 20% to 30% of the sample, stable estimates of its contribution should have been achieved easily. The values in Table 2 however, show a surprising amount of variation.

This variation is depicted graphically in Figure 16 where the standard deviation (s) of the epoxy volume estimates is plotted against the number of sample points. The irregular nature of the lines suggests that the observed variation is virtually independent

Table 2. Results of Replicated Analyses of Epoxy Binder Contribution at Selected Point Densities, for 5 Coals

SAMPLE NO. (PSOC)	SAMPLE POINTS	VOL. % EPOXY	($\bar{x}$) EPOXY %	STANDARD DEVIATION (s)
724	100,000	19.14 18.81 21.51 26.81 19.08	21.07	3.39
724	200,000	19.54 24.67 23.97 21.45 27.39	23.40	3.02
724	400,000	25.97 16.85 19.98 21.10 19.59	20.70	3.34
724	800,000	19.11 21.59 21.33 20.39 21.33	20.75	1.02
773	100,000	21.28 23.46 22.80 25.59 20.09	22.64	2.11
773	200,000	28.79 22.35 21.44 27.32 29.12	25.80	3.65
773	400,000	27.71 23.11 21.52 21.19 24.43	23.59	2.64

Table 2. Results of Replicated Analyses of Epoxy Binder Contribution at Selected Point Densities, for 5 Coals (Continued)

SAMPLE NO. (PSOC)	SAMPLE POINTS	VOL. % EPOXY	($\bar{x}$) EPOXY %	STANDARD DEVIATION (s)
773	800,000	24.10 24.42 28.02 26.86 20.39	24.76	2.94
678	100,000	16.33 16.19 18.41 17.75 17.05	17.15	.94
678	200,000	14.82 17.86 18.70 13.60 14.93	15.98	2.19
678	400,000	15.70 15.62 15.42 18.82 17.52	16.62	1.49
678	800,000	16.72 17.34 16.41 16.82 20.89	17.63	1.85
767	100,000	29.23 26.20 27.97 25.96 27.96	27.46	1.38
767	200,000	24.98 27.52 26.15 28.61 32.31	27.91	2.81

Table 2. Results of Replicated Analyses of Epoxy Binder Contribution at Selected Point Densities, for 5 Coals (Continued)

SAMPLE NO. (PSOC)	SAMPLE POINTS	VOL. % EPOXY	($\bar{x}$) EPOXY %	STANDARD DEVIATION (s)
767	400,000	29.80 24.27 28.77 30.62 24.60	27.61	2.98
767	800,000	24.19 23.05 21.29 22.36 21.66	22.51	1.16
651	100,000	15.74 13.60 7.83 12.21 11.33	12.14	2.93
651	200,000	14.19 12.24 10.58 11.38 12.86	12.25	1.39
651	400,000	16.46 16.88 16.30 13.71 14.11	15.49	1.47
651	800,000	13.89 15.42 19.08 16.10 17.82	16.46	2.03

Figure 16. STANDARD DEVIATION (s) OF EPOXY CONTENT VS LOG NUMBER OF SAMPLE POINTS FOR FIVE SELECTED COALS

of the number of sample points. The expected relationship is the same as was shown in Figure 15 for coals 3, 4 and 5 owing to the inverse exponential relationship between the number of sample points and the sample standard deviation.

The coefficient of variation is the standard deviation expressed as a percentage of the mean ($s/\bar{x}$) and is a useful statistic for comparing the absolute variation of different populations. The data from Tables 1 and 2 are expressed in terms of this value in Table 3. In less than half the cases the pyrite content varies more than the epoxy volume estimates. This means that in an analysis, the epoxy estimate may actually control the precision of the pyrite estimate because its contribution is necessarily subtracted from the total reflectance distribution before the pyrite content is calculated (see Section IIIB).

Investigation of this problem identified three factors contributing to the variation in epoxy volume estimates:

- 1) A minor sensitivity loss of the analog to digital converter in the very low reflectance range, resulting in improper digitized values for the epoxy resin.

- 2) Focusing variation during an analysis severe enough to cause epoxy values to be misplaced within the coal reflectance distribution (and vice versa).

- 3) A varying number of 'edge hits' that occur from analysis to analysis. When the photomultiplier sensing area is occupied by both coal and epoxy, the reflectance value returned is intermediate between the two. Only a 10% areal contribution of coal ($.4 \mu\text{m}^2$) may be enough to misplace the epoxy value in the coal range. The edge effects

Table 3. Comparisons of Pyrite and Epoxy Binder Variation in Replicated Analyses for 5 Selected Coals.

SAMPLE NO. (PSOC)	SAMPLE POINTS	COEFFICIENT OF VARIATION-PYRITE	COEFFICIENT OF VARIATION-EPOXY
724	100,000	.76	.16
	200,000	.35	.13
	400,000	.16	.16
	800,000	.15	.05
773	100,000	.23	.09
	200,000	.11	.14
	400,000	.08	.11
	800,000	.05	.12
678	100,000	.19	.06
	200,000	.06	.14
	400,000	.05	.09
	800,000	.05	.10
767	100,000	.32	.05
	200,000	.13	.10
	400,000	.14	.11
	800,000	.15	.05
651	100,000	.26	.24
	200,000	.23	.11
	400,000	.12	.10
	800,000	.04	.12

become particularly important when the sample contains a large number of 'fines' (under-sized particles).

Unfortunately, within the present analytical capability, these problems cannot be eliminated. Although the use of embedded coal blocks for analysis would circumvent the need for the epoxy binder, many more samples would have to be analyzed in order to obtain a stable estimate of pyrite content for the entire sample. The advantage of a particulate sample is that it represents a large quantity of coal and because of this, the volume estimates stabilize much faster.

The addition of certain dyes to the epoxy would raise its reflectance value and therefore circumvent the improper digitizing in the low reflectance range. The major disadvantage of this technique is that the epoxy contribution must then be deconvoluted from within the coal range where the chances of including data from other populations are greatly increased. A promising approach would be to locate an epoxy resin whose index of refraction is identical to that of the immersion oil used. This would eliminate reflection from the epoxy as light passed the oil/epoxy interface. Theoretically, this would completely remove the epoxy peak from the reflectance distribution of a coal.

The problem of defocusing during an analysis may be mitigated by the addition of an automatic focusing device to the system. Such devices are available commercially, but this source of variation is probably not significant enough to justify the additional expenditure. A trained operator should always be present to adjust the focus during an analysis in order to prevent any major inaccuracies.

Irrespective of the errors inherent in an automated analysis, the results are consistently more reliable than those obtained by conventional means. The major source of error in a conventional analysis involves subjective decision making (operator variation). Further, a trained operator can count only approximately 1000 points in 40 minutes whereas the Rapid Scan can analyze 800,000 points (100 traverses) and produce a hard copy of the results in the same amount of time. In Table 4, the results of five replicated analyses are shown in a comparison of the conventional visual and automated techniques for PSOC-773. In this example, the mean pyrite estimates are very similar, but the standard deviation and 95% confidence intervals of the conventional technique are much larger for the conventional technique. Many times, when determining the amount of a very minor constituent such as pyrite, even 1000 points are not enough to arrive at a stable estimate.

For comparative purposes, the coefficients of variation ($s/\bar{x}$) have been plotted against the pyrite contents of several coals in Figure 17. The estimates stabilize earliest for 800,000 point analyses, and as pyrite content increases, the difference between the point densities decreases. The line for the 100,000 point analyses indicates that this point density (12 traverses) is not suitable for analyzing pyrite in these small percentages. Figure 17 may be used to help determine the appropriate sampling plan for a given level of precision, or conversely, to estimate the level of precision for a single analysis.

Another factor that influences the precision of the volumetric estimates is the nature of the pyrite size distribution on the sample

Table 4. Comparison of Pyrite Volume Estimates from Automated and Conventional Techniques for PSOC-651

<u>AUTOMATED</u>		<u>CONVENTIONAL</u>
800,000	SAMPLE POINTS	1,000
.89		.90
.94		1.20
.98	VOLUME %	.30
.90	ESTIMATES	.70
.95		1.80
	MEAN PYRITE	
.93	CONTENT ($\bar{x}$)	.98
.04	STANDARD DEVIATION (s)	.56
$.90 \leq \mu \leq .96$	95% CONFIDENCE LIMITS	$.50 \leq \mu \leq 1.46$

μ = the population mean or the 'true' amount of pyrite in the coal sample.

Figure 17. COEFFICIENT OF VARIATION($s/\bar{x}$)VS PYRITE CONTENT FOR FIXED NUMBERS OF SAMPLE POINTS IN A RAPID SCAN AUTOMATED ANALYSIS

surface. The repeated analysis of a sample with 1% pyrite by volume would show a large variation if the pyrite is contained in a few large blebs. The inclusion or exclusion of one chord has a proportionally large influence on the estimate. If the same amount of pyrite is dispersed as much smaller particles on the sample surface, each chord would therefore represent a smaller portion of the total volume. An increase in the potential number of targets entails a corresponding increase in the precision of replicated analyses.

The interaction of the sampling grid with the pyrite size distribution is depicted in Figure 18. Here, the number of pyrite chords encountered for every 100,000 sample points is plotted against the total number of points in the analysis. In the case of coal 2, with each increase in point density, the number of pyrite particles encountered also increases. This indicates that as the sample lines are moved closer together on the specimen surface, more and more small particles are being intersected. Hence, the volume estimates should also become more stable. This is borne out by the curve for coal 2 that appeared in Figure 15. Recall that the standard deviation of pyrite volume estimates showed a continual decrease and that there was no inflection point in this curve indicating that an increase in point density even beyond 800,000 may have been warranted. Also in Figure 17, coals 3 and 4 show constant values for each 100,000 points and this is also manifest in Figure 15, where the curves closely approximate the expected relationship.

Because it is impossible to estimate the amount and distribution of pyrite in a sample prior to an analysis, it is always advisable to use the maximum point density consistent with the constraints of time.

Figure 18. NUMBER OF PYRITE PARTICLES PER 100,000 SAMPLE POINTS VS THE TOTAL NUMBER OF SAMPLE POINTS IN AN AUTOMATED ANALYSIS

This is especially true when only one or two analyses are required.

However, if the situation warrants the analysis of many samples from the same pyrite population, then the kind of information in Figures 15, 17 and 18 will be useful in determining the most efficient sampling plan.

B. The Effect of Sample Variation on the Volume Estimates of Pyrite Content in a Coal

At The Pennsylvania State University Coal Research Section, the routine preparation of particulate coal samples for petrographic analysis yields a set of 6 polished pellets. For one coal, PSOC-773, these pellets were tested for non-randomness of pyrite distribution. Each sample was analyzed 4 times at a point density of 400,000. The results of these 24 analyses are summarized in quality-control form in Figure 19. The grand mean ($\bar{\bar{x}}$) for the six pellets was determined to be .49% pyrite and was plotted on the graph. The 95% confidence limits ($s = .12$) then were added as dashed lines. Mean values for each sample (4 replicates) were then calculated and plotted and points that fell outside of the $\pm 2s$ confidence limits were considered to be out of control (they have pyrite contents significantly different than would be expected of random samples from the same population). Three of the six pellets were found to be non-random samples.

Pellet 1 ($\bar{x} = .39\%$) and pellet 6 ($\bar{x} = .38\%$) contained too little pyrite and pellet 4 ($\bar{x} = .67\%$) contained too much. Pellets 2 ($\bar{x} = .50\%$), 3 ($\bar{x} = .48\%$) and 5 ($\bar{x} = .53\%$) all fell within the

Figure 19. QUALITY CONTROL GRAPH OF GRAND MEAN ($\bar{\bar{x}}$) OF PYRITE CONTENT AND 95% CONFIDENCE LIMITS VS SAMPLE MEANS ($\bar{x}$) FOR PSOC-773

acceptable range. These results indicated that sampling, sample preparation, or both are significant problems in the pyrite analysis. Whatever the cause of this unacceptable variation in the amount of pyrite, it is clear that more than one pellet (3 in this case) should be analyzed, and a mean value used in further computations.

A similar treatment of the data for the three acceptable pellets is shown in Figure 20. The grand mean for these 3 pellets was calculated ($\bar{x} = .50\%$) as were the 95% confidence limits ($s = .04$). In this case, pyrite estimates were chosen randomly from the 4 replicated analyses for each sample. First, only one analysis was selected from each set and plotted against the grand mean. Next, 2 analyses were chosen, their mean calculated, and plotted. This process was repeated for 3 and 4 analyses on each pellet. Figure 20 shows that for 2 of the 3 pellets, mean values from 2 selected analyses fall beyond the confidence limits. For pellet 3, the estimate is higher than was expected, and for pellet 5 it is lower. Note that for all three pellets, the mean values for both 3 and 4 replicates give acceptable estimates, and that there is very little difference between the two. The results of this study indicate that, for this coal, at least 3 replicate analyses must be made on a random sample before an acceptable estimate of pyrite content can be assured.

Figure 20. QUALITY CONTROL GRAPH FOR GRAND MEAN ($\bar{\bar{x}}$) OF PYRITE CONTENT FROM 3 RANDOM SAMPLES OF PSOC-773 VS SAMPLE MEANS ($\bar{x}$) FROM A SELECTED NUMBER OF REPLICATED ANALYSES ON EACH

V. THE PYRITE CHORD-SIZE DISTRIBUTION

A. Parametric Statistical Approach

Upon completion of the volumetric pyrite analysis, the Rapid Scan also provides a distribution of pyrite chord lengths. This distribution, in histogram form, is divided into classes for every 2 μm length. The typical shape of this distribution is illustrated in Figure 21. The chord length values have a potential range of 2 μm (the limit of resolution for the system) to about 850 μm , which is the diameter of the opening in a 20 mesh sieve used in the standard sample preparation (ASTM D2797-72, 1978). These distributions always have a high positive skew (mean greater than the mode) owing to the fact that most measurements occur in the smallest size classes, near the resolving limit of the system.

Because much statistical inference is based on the assumption that random samples have been drawn from a normally distributed population, selected data transformations were performed in order to improve the quality of the chord size data in this regard. The program NORMSTAT, written for the IBM 370 computer, was used to transform the data and fit the resulting distribution to the expected normal curve. In Figure 22, both the observed and expected pyrite chord length distributions are plotted for untransformed data from sample PSOC-773. The class intervals are based on fractions of the sample standard deviation (s). Again, note the excess of readings (observed greater than expected) in the smallest size classes, and the complete absence of readings beyond this point (below the resolving limit). The summary statistics of skewness ($\sqrt{b_1}$), kurtosis (b_2) and chi squared

CHORD SIZE(UM)	CUM %	FREQUENCY							
		5 V	10 V	15 V	20 V	25 V	30 V	35 V	
-2	11.02	*****							
4	20.47	*****							
6	30.71	*****							
8	39.37	*****							
10	45.67	*****							
12	50.39	*****							
14	54.33	*****							
16	58.27	*****							
18	62.99	*****							
20	63.78	*							
24	66.14	***							
26	66.93	*							
28	70.08	****							
30	73.23	****							
32	75.59	***							
34	78.74	****							
36	80.31	**							
38	81.89	**							
42	83.46	**							
48	85.04	**							
50	85.83	*							
52	86.61	*							
54	88.19	**							
56	88.98	*							
64	89.76	*							
72	90.55	*							
76	92.13	**							
82	92.91	*							
84	93.70	*							
98	94.49	*							
104	95.28	*							
110	96.06	*							
122	97.64	**							
132	98.43	*							
134	99.21	*							
204	100.00	*							

Figure 21. A HISTOGRAM OF PYRITE CHORD LENGTHS

Figure 22. UNTRANSFORMED PYRITE CHORD-SIZE DISTRIBUTION FOR PSOC-773

(χ^2) are all highly significant, indicating that, in the 'as received' state, the pyrite chord size distribution does not fit the expected normal model.

The data transformations were chosen for their ability to reduce the skewness and enhance the central tendency of the data. In Figure 23, the result of a square root transformation on the PSOC-773 data is shown. Here the shape of the distribution is somewhat changed, but the highly significant summary statistics indicate rejection of the normal model. The same situation exists for the arcsine transformation shown in Figure 24, and the hypothesis of random samples from a normally distributed population must again be rejected.

As some size distributions found in nature are known to be log-normal (Krumbein (1938), Gaddum (1945), and Griffiths (1967, Ch. 5)), three different transformations involving logarithms were tried with the PSOC-773 data. The results for all three were virtually identical, so only the $\log_{10}$ transformation is shown in Figure 25. While this transformation appears to give the best result, the kurtosis (b_2) and chi squared (χ^2) are still highly significant. The skewness parameter ($\sqrt{b_1}$) is not significant at the 5% level, however.

The results of these transformations are summarized graphically in quality control form in Figure 26. The 5% and 1% confidence levels for skewness and kurtosis define a rectangular target area of acceptability in the graph. Points that fall beyond this central rectangle are considered to be out of control. The untransformed distribution lies the farthest distance from the target, while both the arcsine and square root transformations show considerable improvement. The best results are obtained for the logarithmic

Figure 23. SQUARE ROOT TRANSFORMATION OF PYRITE CHORD-SIZE DISTRIBUTION FOR PSOC-773

Figure 24. ARCSINE TRANSFORMATION OF PYRITE CHORD-SIZE DISTRIBUTION FOR PSOC-773

Figure 25. LOG_{10} TRANSFORMATION OF PYRITE CHORD-SIZE DISTRIBUTION FOR PSOC-773

Figure 26. b_2 VS $\sqrt{b_1}$ TEST FOR NONRANDOMNESS OF PYRITE CHORD-SIZE DISTRIBUTION FOR PSOC-773

transformations, but the low value for kurtosis keep the points from falling into the acceptable region. In conclusion, the results from this suite of pyrite chord-size analyses for PSOC-773 caused the hypothesis of random samples from a normally distributed population to be rejected.

These transformations were also performed on three other coals whose summary statistics appear in Table 5. These results, in quality control form, appear in Figure 27. The sample size for each distribution appears in the upper left hand corner of each graph. Generally, for all three coals, the effectiveness of a transformation was found to be a function of sample size (the number of chords in the distribution). As the sample size increased, the untransformed data points and square root data points moved farther from the acceptable region. The effect of the arcsine transformation was variable, and was found to be unsuitable for the chord-size distribution of PSOC-724 because of the small sample size. The logarithmic transformation was able to reduce skewness in all but the largest sample (PSOC-678), and most often gave the best results. Importantly, none of these operations yielded a distribution consistent with the normal model indicating that another approach was necessary in order to effectively compare these distributions to one another.

B. Nonparametric Statistical Approach

The two aspects of statistical inference entail the estimation of population parameters and the testing of various hypotheses based on a limited sample size. In the parametric analysis of pyrite chord-length distributions, it is assumed that these population parameters

Table 5. Pyrite Chord-Size Distribution Summary Statistics for Samples PSOC-678, PSOC-706 and PSOC-724.

PSOC-678	PSOC-706	PSOC-724
<u>Untransformed</u>	<u>Untransformed</u>	<u>Untransformed</u>
n = 287 μ m	n = 169 μ m	n = 28 μ m
$\bar{x}$ = 28.56 μ m	$\bar{x}$ = 30.17 μ m	$\bar{x}$ = 61.43 μ m
s = 31.81 μ m	s = 45.24 μ m	s = 74.83 μ m
$\sqrt{b_1}$ = 4.08**	$\sqrt{b_1}$ = 3.77**	$\sqrt{b_1}$ = 1.52**
b ₂ = 20.52**	b ₂ = 22.36**	b ₂ = 4.35**
χ^2_{18} = 704.56**	χ^2_{16} = 401.13**	χ^2_{10} = 42.84**
<u>Square Root</u>	<u>Square Root</u>	<u>Square Root</u>
n = 287	n = 169	n = 28
$\bar{x}$ = 16.36 μ m	$\bar{x}$ = 20.88 μ m	$\bar{x}$ = 42.25 μ m
s = 11.82 μ m	s = 9.04 μ m	s = 19.90 μ m
$\sqrt{b_1}$ = 2.59**	$\sqrt{b_1}$ = 1.56**	$\sqrt{b_1}$ = .74**
b ₂ = 10.29**	b ₂ = 6.20**	b ₂ = 2.53
χ^2_{18} = 328.42**	χ^2_{16} = 104.01**	χ^2_{10} = 18.05*
<u>Log</u>	<u>Log</u>	<u>Log</u>
n = 287	n = 169	n = 28
$\bar{x}$ = 10.82 μ m	$\bar{x}$ = 14.02 μ m	$\bar{x}$ = 25.73 μ m
s = 3.45 μ m	s = 3.50 μ m	s = 4.53 μ m
$\sqrt{b_1}$ = .82**	$\sqrt{b_1}$ = .19	$\sqrt{b_1}$ = -.161
b ₂ = 3.43**	b ₂ = 2.18**	b ₂ = 1.80**
χ^2_{18} = 155.56**	χ^2_{16} = 87.75**	χ^2 = 14.09
<u>Arcsine</u>	<u>Arcsine</u>	
n = 287	n = 169	
$\bar{x}$ = 14.14 μ m	$\bar{x}$ = 14.38 μ m	
s = 14.27 μ m	s = 14.04 μ m	
$\sqrt{b_1}$ = -1.12**	$\sqrt{b_1}$ = -1.34**	
b ₂ = 3.18	b ₂ = 3.73**	
χ^2_{18} = 851.69**	χ^2_{16} = 309.53**	

* = significant at 5% level of probability

** = significant at 1% level of probability

Figure 27. b_2 VS $\sqrt{b_1}$ TEST FOR NON-RANDOMNESS OF PYRITE CHORD-SIZE DISTRIBUTION AT 95% CONFIDENCE LEVEL FOR 3 SELECTED COALS

have certain values. Since the points in Figures 26 and 27 lie beyond the acceptable range for these values (at a specified probability level), the hypothesis of a normal model must be rejected. In nonparametric, or distribution-free statistics, conditions about the parameters of the population from which the samples are taken are not specified. Hence, conclusions concerning differences between sample sets are drawn without benefit of a model and the derived probability statements are independent of the shape of the population (Siegel, 1956, Ch. 3)). These statistics can be of great value when little is known about the population being sampled or, as in the case of the pyrite chord distribution, it is necessary to be free of the requirements of the various parameteric tests.

The Kolmogorov-Smirnov two-sample test is a nonparametric statistic that is sensitive to all changes in the distributions from which the samples are drawn (Ibid, Ch. 6). A large enough difference in central tendency or skewness will result in rejection of the hypothesis that both samples are drawn from the same population distribution. Although the statistic cannot discriminate the nature of the difference, it is still very useful in quality control situations where it is necessary only to know that this difference exceeds some acceptable limit. On the other hand, if two samples are drawn from the same population, only minor random variations should be observed and the magnitude of any differences should be relatively small (Ibid, Ch. 5).

The statistic is based on a comparison of the cumulative frequency distributions for each set of observations as follows:

$$A_1(L) = K/n_1 \text{ and}$$

$$A_2(L) = K/n_2 \text{ where}$$

$A_1(L)$, $A_2(L)$ = the observed cumulative step function of the samples, K = the number of scores less than or equal to L , and n_1 and n_2 are the sample sizes of the two observed distributions. In the pyrite chord size distribution, L increases in size by increments of 2 μm .

The difference value (D) is then equal to:

$$D = \text{maximum}|A_1(L) - A_2(L)|$$

This value is tested for significance against tabled values which were calculated from the sampling distribution of D , at various probability levels. Thus, the D value provides a means of testing the hypothesis of whether two pyrite chord-size distributions have been drawn from the same population distribution and it has been used routinely to compare the Rapid Scan analyses.

VI. APPLICATION OF THE RAPID SCAN AUTOMATED PYRITE ANALYSIS TO COAL BENEFICIATION

In order to study the effects of comminution on the pyrite content of a coal, a 10-pound sample of PSOC-634, the Upper Freeport seam, was crushed to pass an 8-mesh (U.S. Standard) sieve. This size range approximates that of a fine-coal cleaning circuit in a coal preparation plant. Subsamples were taken from the 8 x 0 mesh range, pelletized, and analyzed by the Rapid Scan for their volume percent pyrite (1.82%) and mean pyrite chord length (16.2 μm). The mean chord length was computed from log-transformed data and should be regarded with caution, owing to the non-normality of the data.

Next, the bulk of the coal sample was separated into its natural size distribution using a series of 21 closely sized sieves. Each individual fraction was then analyzed by the Rapid Scan for its pyrite content, and these results were compared to those obtained from the composite sample. The pyrite chord-size distribution for each fraction was compared to that obtained from the next larger fraction to test whether a significant change had taken place. This was determined by testing the D-value from the Kolmogorov-Smirnov test statistic. The results of these analyses are shown in Table 6. Two pellets were analyzed from each fraction, and their mean values reported in the Table. With this information, it was possible to identify fractions containing anomalously high concentrations of pyrite, as well as those which contained different size distributions of pyrite.

Table 6. Automated Analysis of Specially Sized Suite for PSOC-634.

<u>FRACTION</u>	<u>OPENING</u>	<u>$\bar{x}$ CHORD</u>	<u>% PYRITE</u>	<u>D - VALUE</u>
8 x 0	2.36 mm	16.2 μ m	1.82	--
8 x 10	2.36 mm	13.6 μ m	1.42	.076*
10 x 12	2.00 mm	18.3 μ m	3.83	.086**
12 x 14	1.70 mm	11.7 μ m	1.51	.148***
14 x 16	1.40 mm	15.8 μ m	2.03	.105***
16 x 18	1.18 mm	14.7 μ m	1.24	.055
18 x 20	1.00 mm	15.4 μ m	1.65	.046
20 x 25	850 μ m	17.2 μ m	1.29	.051
25 x 30	710 μ m	14.0 μ m	1.46	.087*
30 x 35	600 μ m	16.5 μ m	1.65	.075*
35 x 40	500 μ m	13.4 μ m	1.71	.103***
40 x 45	425 μ m	13.8 μ m	.90	.072*
45 x 50	355 μ m	17.1 μ m	1.50	.112**
50 x 60	300 μ m	12.3 μ m	.92	.154***
60 x 70	250 μ m	12.0 μ m	1.01	.046
70 x 80	212 μ m	11.5 μ m	.82	.040
80 x 100	180 μ m	11.8 μ m	.79	.020
100 x 120	150 μ m	13.0 μ m	1.06	.060
120 x 170	125 μ m	11.3 μ m	1.19	.068*
170 x 200	90 μ m	9.0 μ m	1.16	.096***
200 x 270	75 μ m	7.0 μ m	1.28	.117***
270	53 μ m	5.5 μ m	.65	.142***

* - SIGNIFICANT AT 5% LEVEL OF PROBABILITY

** - SIGNIFICANT AT 1% LEVEL OF PROBABILITY

*** - SIGNIFICANT AT .1% LEVEL OF PROBABILITY

From the coarsest to finest size fractions, the first significant changes occur in the 8 x 16 mesh range, which comprises 4 size fractions. Each fraction in this range contains a different pyrite chord-size population. The 10 x 12 mesh fraction contains both the highest proportion of pyrite (3.83%) and the largest mean chord length (18.3 μm) of any fraction. The 14 x 16 mesh fraction also had a slightly higher pyrite content than the composite (8 x 0) sample. These changes indicate that the largest of the pyrite particles had begun to interact with the crushing operation. Since the first episode of pyrite change ends with the 14 x 16 mesh fraction, a screening operation and density separation at this point in the preparation facility would be most effective.

After 3 fractions of no further change, the pyrite size distribution shows a significant difference in the 25 x 30 mesh fraction. In each of the next 5 fractions, the chord-size distribution also differed significantly. In these cases, there are no anomalous pyrite concentrations, and in fact, the 40 x 45 and 50 x 60 mesh fractions contain less pyrite than expected, based on the composite (8 x 0) sample.

The changes in the chord-size distributions indicate that the relationship of the pyrite particles to the surrounding coal had changed, and presumably, some more pyrite had been 'liberated' (liberated, as used here, means that the ratio of pyrite to coal in a particle has become sufficiently high to allow the particle to be removed in a density separation). Thus, another separation should be implemented in this mesh range, perhaps with a cyclone, or other technique capable of handling these small sizes.

A final episode of pyrite size change occurs in the four smallest size fractions. Note that the mean pyrite chord length shows a decrease with each successive comminution. At these very small sizes, most of the particles reacted to the crushing and consequently reflect this trend in the mean chord-size values. Significantly, in these fractions there are no anomalously high pyrite concentrations. If a coal is crushed excessively, a disproportionate number of fine particles can be created. These particles tend to decrease the efficiency of any gravity separation process and for the most part, they survive the entire cleaning circuit relatively unaffected. Therefore, the pyrite that occurs in these fractions will eventually be combusted.

In summary, there were observed three prominent episodes of pyrite particle size change in the size fractionation of this coal. The first occurred in the larger sizes as a result of the comminution of the largest pyrite particles. A gravity separation was indicated at the 14 x 16 fraction in order to take advantage of these changes and the anomalously high pyrite concentrations in two of the fractions. The second episode of pyrite size change occurred in the intermediate size fractions as a response of the natural distribution of pyrite sizes to the crushing operation. In this case a gravity separation was indicated in the 50 x 60 mesh fraction. The final size changes occurred in the smallest increments where virtually every particle responded to further comminution.

Three important conclusions drawn from the preceeding demonstration of the use of automated analysis will lead to the eventual application of the Rapid Scan system to a real coal cleaning situation. They are:

- 1) The ability of the Rapid Scan system to characterize many samples of a feedcoal quickly and precisely.
- 2) The ability of the Rapid Scan system to locate size fractions of high pyrite concentration.
- 3) The ability of the Rapid Scan system to identify significant changes in pyrite chord size distribution.

Sophisticated coal cleaning plants operate their fine-coal circuits in several size ranges. Knowledge of the nature of the coal substance could be used to modify these circuits, and thus increase the overall efficiency of the cleaning operation. Several studies have been made (Benedict (1962), Bayer (1960), Shapiro et al. (1969)) concerning the response of the organic constituents of coals to various cleaning and sizing operations. Now, with the capabilities of the Rapid Scan system of automated reflectance microscopy, there is no reason why similar studies could not be undertaken with pyrite, the principal sulfur bearing mineral of coal. The prospects of the more efficient utilization of indigenous energy sources, should be stimulus enough to initiate large-scale, practically oriented research with this equipment.

VII. CONCLUSIONS AND SUGGESTIONS FOR FURTHER RESEARCH

1) There are numerous systems for automated image analysis in use today. The 'image-plane' or television-type scanners can provide a variety of pattern and shape data, but are generally hardware reliant and possess a number of major problems in the detection of features. 'Specimen-plane' scanners are free from most of these problems, but are capable only of basic lineal analysis (Section IA).

2) The earliest attempts to automate various microscopic coal analyses began over 20 years ago, but were limited by the relatively crude instrumentation available at the time. Two important problems were the proper demarcation of particle boundaries and the treatment of gaps and discontinuities within particles (Section IB).

3) To a certain extent, analytical capability is a function of system cost. The Pennsylvania State University Rapid Scan system utilizes a software base and conventional hardware components in order to achieve flexibility at a relatively low total cost (Section IIA).

4) In order to calculate the volume of pyrite in a sample with the Rapid Scan system, the reflectance distribution of the coal (in oil) in the range of 0-8% is stored in computer memory. The readings on pyrite are always stored in the last array element (1024). After the contribution of the epoxy binder in the sample has been subtracted, the ratio of the number of readings on pyrite to the remaining readings yields the volume estimate of pyrite (Section IIIB).

5) In the analysis of pyrite volumes, the estimation of the epoxy binder was shown to exhibit greater variation than the pyrite estimates. Hence, precision was controlled by the inability to

effectively remove readings from the low reflecting epoxy. Three sources of this variation were identified:

- a) a minor sensitivity loss in the analog to digital converter of the computer in the low reflectance range.

- b) the occurrence of focusing inaccuracies during an analysis.

- c) a variable number of 'edge hits' from analysis to analysis (Section IVA).

6) Despite these errors, the Rapid Scan estimates are much more precise than those derived from the conventional point counting techniques (Section IVA).

7) In a study of the effect of sample variation on pyrite volume estimates, 3 of the 6 samples were found to be non-random. This indicated that in order to attain the maximum accuracy, several samples of each coal should be analyzed. It was also shown that as many as 3 replicate analyses were necessary to attain the maximum precision possible (Section IVB).

8) A modification of the Rosiwal analysis allows a distribution of chord-size measurements to be accumulated during a Rapid Scan analysis. The complex algorithm has four branches of unequal length which must take the same real amount of execution time so that the automatic stage drive maintains the proper spatial relationship between consecutive readings (Section IIIC).

9) The significant features of the pyrite chord size distribution were found to be:

- a) a high positive skew (mean greater than the mode) with,

b) the majority of readings occurring near the resolving limit of the system.

In the analysis of these distributions, a parametric statistical approach was rejected because the data did not become normal (Gaussian) after a series of transformations was applied (Section VA).

10) A nonparametric test, The Kolmogorov-Smirnov statistic was adopted for the routine comparison of chord-size distributions. Although the test cannot elucidate the nature of the difference between two samples, it does indicate whether or not they have been drawn from the same population (Section VB).

11) The analytical capabilities of the Rapid Scan were applied in a simulation of the crushing operations in the fine-cleaning circuit of a coal preparation plant. Three episodes of pyrite size distribution change were noted in the 8 x 16, 25 x 60 and 170 x 270 mesh ranges. Also, two fractions were shown to have significantly high pyrite concentrations (Section VIA). With this information, it may be possible to predict where the most efficient gravity separations can be made in a preparation plant, thus maximizing the removal of pyrite from a given feed coal (Section VIA).

Within the cadre of the present Rapid Scan analytical capability, the developmental aspects of this research have been completed. It remains now for this technique to be applied in the appropriate situations. For example, correlation of these data to actual float-sink data from various coal cleaning techniques would further enhance the benefit of this equipment to coal preparation.

The ability to resolve particles below the present 2 μ m limit would be of great value in any further development of Rapid Scan

hardware. This could conceivably change the shape of the chord-size distribution and make it amenable to improved statistical treatment. Thus, further improvements in the Rapid Scan data will be directly related to increases in the analytical capability of the system.

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APPENDIX A

Basic FORTRAN Data Acquisition Program for the Rapid Scan
Automated Pyrite Analysis

```

C                                CHORD.FT
C
C
C      THIS PROGRAM GENERATES A PYRITE CHORD SIZE DISTRIBUTION
C      IT COMPUTES THE VOL% PYRITE AND SUMMARY STATISTICS
C
C      KENNETH W. KUEHN                      2/10/77
C
C      THIS SECTION STANDARDIZES THE SYSTEM
COMMON ARR(3050),JFAKE(4),JREF(1024)
INTEGER JSIZE(3000)
EQUIVALENCE (ARR(51),JSIZE(1))
WRITE(4,901)
901  FORMAT(' ENTER REFLECTANCE VALUE FOR GLASS STANDARD      ',5)
      READ(4,902)RFS
902  FORMAT(F9.4)
900  CALL SETADC(32,0)
      DO 915 J=1,3
      WRITE(4,915)
915  FORMAT(1X)
      WRITE(4,916)
916  FORMAT(' THE FULL SCALE REFLECTANCE VALUE IS: ')
      DO 905 J=1,19
      CALL FASTAD(8,JSS)
      CALL FASTAD(8,JS2)
      CALL FASTAD(8,JS3)
      CALL FASTAD(8,JS4)
      CALL FASTAD(8,JS5)
      CALL FASTAD(8,JS6)
      RSS=FLOAT(JSS)+FLOAT(JS2)+FLOAT(JS3)
      RS1=FLOAT(JS4)+FLOAT(JS5)+FLOAT(JS6)
      RSS=(RSS+RS1)/6.
      RD=1000./RSS
      RE=RD*RFS
      WRITE(4,910)RE
910  FORMAT(1X,F8.3)
      DO 908 JA=1,5000
908  CONTINUE
905  CONTINUE
      WRITE(4,920)
920  FORMAT(' DO YOU WANT TO REPEAT?',5)
      READ(4,940)J1
940  FORMAT(A1)
      IF(J1.EQ.'Y')GO TO 900
C      ESTABLISHES RUN PARAMETERS
DO 1 JD=1,1024
1    IREF(JD)=0
DO 2 JE=1,3000
2    JSIZE(JE)=0
DO 3 JA=1,4
3    JFAKE(JA)=0
DO 7 JZ=1,50
7    ARR(JZ)=0.0
      WRITE(4,201)
201  FORMAT(' ENTER DATE IN 6 CHARS. EG. 27-MAR      ',5)
      READ(4,202)ARR(1)
202  FORMAT(A6)
      WRITE(4,203)
203  FORMAT(1X)

```

```

204 WRITE(4,204)
FORMAT(' ENTER PSOC NUMBER. EG. 9999', $)
READ(4,205)JSJD
205 FORMAT(I5)
ARR(4)=FLOAT(JSJD)
WRITE(4,206)
206 FORMAT(1X)
WRITE(4,215)
215 FORMAT(' ENTER ANY SPECIAL JD. MAX JS 10 CHARS.', $)
READ(4,216)ARR(2),ARR(3)
216 FORMAT(2A6)
WRITE(4,217)
217 FORMAT(1X)
WRITE(4,207)
207 FORMAT(' ENTER PHOTOMULTIPLIER SPOT SIZE', $)
READ(4,208)JSPOT
208 FORMAT(I2)
ARR(5)=FLOAT(JSPOT)
WRITE(4,209)
209 FORMAT(1X)
WRITE(4,211)
211 FORMAT(' ENTER STAGE DRIVE SPEED IN MICRONS/SEC', $)
READ(4,212)ISPD
212 FORMAT(I4)
ARR(6)=FLOAT(ISPD)
WRITE(4,213)
213 FORMAT(1X)
WRITE(4,214)
214 FORMAT(' ENTER NUMBER OF READINGS PER TRAVERSE', $)
READ(4,20)ITRAV
20 FORMAT(I6)
WRITE(4,26)
26 FORMAT(1X)
WRITE(4,30)
30 FORMAT(' ENTER TOTAL NUMBER OF READINGS DESIRED', $)
READ(4,40)JTOT
40 FORMAT(I8)
ARR(6)=FLOAT(JTOT)
TLEN=(ARR(5)*ARR(6))/1000.
ARR(7)=TLEN
WRITE(4,45)
45 FORMAT(1X)
WRITE(4,50)
50 FORMAT(' ENTER LENGTH OF DISCONTINUITY TO BE BRIDGED', $)
READ(4,60)JGAP
60 FORMAT(I3)
ARR(9)=FLOAT(JGAP)
JBRJD=JGAP/JSPOT
WRITE(4,65)
65 FORMAT(1X)
WRITE(4,70)
70 FORMAT(' ENTER DELAY FACTOR', $)
READ(4,80)IDEL
80 FORMAT(I4)
WRITE(4,85)
85 FORMAT(1X)
WRITE(4,90)
90 FORMAT(' ***** TURN ON STAGE DRIVE ***** ')
C DATA ACQUISITION BEGINS HERE
WRITE(4,95)
95 FORMAT(1X)
6 CALL SETADC(32,1)
CALL FASTAD(6,KEN)

```

```

IF (KEN.GT.500)GO TO 6
CONTINUE
NTOT=0
JFLAG=0
JPART=0
NTRAV=0
KNTTR=1
GO TO 11
200 DO 4 L=1,JDEL
4 CONTINUE
NTOT=NTOT+1
IF (NTOT.GT.JTOT)GO TO 15
NTRAV=NTRAV+1
IF (NTRAV.GT.JTRAV)GO TO 13
CALL FASTAD(3,JVAL)
JREF(JVAL)=JREF(JVAL)+1
IF (JVAL.GT.999)GO TO 5
IF (JFLAG.EQ.0)GO TO 25
IF (K.GT.JBRID)GO TO 8
K=K+1
KDUM=0
JDUM=2
JDUM=0
JFAKE(JDUM)=JFAKE(JDUM)+1
IF (JDUM.GT.3000)JDUM=3000
GO TO 200
8 IF (JCHOR.GT.3000)JCHOR=3000
JSJZE(JCHOR)=JSJZE(JCHOR)+1
JPART=JPART+1
K=0
JCHOR=0
JFLAG=0
GO TO 200
5 IF (JFLAG.EQ.0)JFLAG=1
IF (K.GT.0)JCHOR=JCHOR+K
K=0
JCHOR=JCHOR+1
JDUM=3
JDUM=0
JFAKE(JDUM)=JFAKE(JDUM)+1
IF (JDUM.GT.3000)JDUM=3000
GO TO 200
25 CONTINUE
KDUM=0
JDUM=1
JDUM=0
JFAKE(JDUM)=JFAKE(JDUM)+1
IF (JDUM.GT.3000)JDUM=3000
GO TO 200
C THIS SECTION READS END OF SCAN SIGNAL AND BEGINS NEW TRAVERSE
11 CONTINUE
KNTST=0
13 CONTINUE
CALL SETADC(32,1)
WRITE(4,31)KNTST
31 FORMAT( ' END OF DATA ACQUISITION FOR TRAVERSE NUMBER ',J4)
KNTST=KNTST+1
IF (JFLAG.GT.0)JFLAG=0
K=0

```

```

      JCHOR=0
24    CALL FASTAD(6,JEND)
      IF (JEND.GT.500)GO TO 9
      GO TO 24
9     CONTINUE
      WRITE(4,33)
33    FORMAT(1X)
      WRITE(4,32)
32    FORMAT(' Y MOVEMENT IN PROGRESS',5)
21    CALL FASTAD(6,JEND)
      IF (JEND.GT.500)GO TO 21
      DO 12 JG=1,8000
12    CONTINUE
      WRITE(4,34)
34    FORMAT(1X)
      WRITE(4,22)KNTTR
22    FORMAT(' NOW TAKING DATA ON TRAVERSE NUMBER ',J4)
      KNTTR=KNTTR+1
      NTRAV=0
      CALL SETADC(32,0)
      GO TO 200
15    WRITE(4,210)
210   FORMAT(' *****      RUN COMPLETED      ***** ')
C     CALCULATION OF SUMMARY STATISTICS
      JEPOX=0
      JMAX=0
      DO 766 J=2,20
      JVAL=JREF(J)
      IF (JVAL.GT.JMAX)GO TO 744
      GO TO 766
744   JMAX=JVAL
      JMEAN=J
766   CONTINUE
      DO 733 J=2,JMEAN
      JEPOX=JEPOX+JREF(J)
733   CONTINUE
      JEPOX=(JEPOX*2)+JREF(1)
      EPCT=(FLOAT(JEPOX)/FLOAT(JTOT))*100.
      ARR(48)=EPCT
      COAL=((100.-EPCT)/100.)*FLOAT(JTOT)
      JPY=0
      DO 755 J=1000,1024
      JPY=JPY+JREF(J)
755   CONTINUE
      PYPCT=(FLOAT(JPY)/COAL)*100.
      ARR(49)=PYPCT
      CONTINUE
      JPART=0
      JHJTS=0
      DO 300 J=1,3000
      JPART=JPART+JSIZE(J)
      JHJTS=JHJTS+((JSPOT*J)*JSIZE(J))
300   CONTINUE
      ARR(10)=FLOAT(JPART)
      ARR(14)=FLOAT(JHJTS)
      MJN=1
302   IF (JSIZE(MJN).EQ.0)GO TO 301
      MJN=MJN*JSPOT
      ARR(12)=FLOAT(MJN)
      GO TO 303
301   MJN=MJN+1
      GO TO 302
303   MAX=3000

```

```

306      JF(JSIZE(MAX).EQ.0)GO TO 304
        MAX=MAX*JSPOT
        ARR(13)=FLOAT(MAX)
        GO TO 305
304      MAX=MAX-1
        GO TO 306
305      CONTINUE
C      CALCULATION OF DECILE STATISTICS
        JX=1
        JSUB=0
        KDEC=19
        DO 310 IDEC=1,10
          JPCT=(JHITS*IDEC)/10
          DO 311 J=JX,3000
            JSUB=JSUB+((JSPOT*J)*JSIZE(J))
            IF(JSUB.GE.JPCT)GO TO 312
311          CONTINUE
312          JF(JSIZE(J).EQ.0)GO TO 313
            GO TO 314
313          ARR(KDEC)=ARR(KDEC-1)
            GO TO 315
314          ARR(KDEC)=FLOAT(I*JSPOT)
315          KDEC=KDEC+1
            JX=J+1
310          CONTINUE
            KPART=0
            KDEC=31
            DO 320 J=1,2000
              KPART=KPART+JSIZE(J)
              J=J*JSPOT
              DO 322 J1=10,50,10
                IF(J.EQ.J1)GO TO 321
322              CONTINUE
                IF(J.EQ.100)GO TO 321
                IF(J.EQ.200)GO TO 321
                IF(J.EQ.500)GO TO 321
                IF(J.EQ.1000)GO TO 321
                IF(J.EQ.2000)GO TO 321
                GO TO 320
321              ARR(KDEC)=(FLOAT(KPART)/FLOAT(JPART))*100.
                KDEC=KDEC+1
320              CONTINUE
C      CALCULATION OF MEAN CHORD SIZE FROM LOG10 TRANSFORMED DATA
        GRAND=0.0
        DO 400 J=1,3000
          IF(JSIZE(J).EQ.0)GO TO 400
          SJZE=(ALOG10(FLOAT(J)*ARR(5)))*FLOAT(JSIZE(J))
          GRAND=GRAND+SJZE
400        CONTINUE
          AMEAN=GRAND/ARR(10)
          CMEAN=10**AMEAN
          ARR(41)=CMEAN
C      CALCULATION OF STANDARD DEVIATION
        GDJFF=0.0
        DO 410 J=1,3000
          IF(JSIZE(J).EQ.0)GO TO 410
          SDJFF=((ALOG10(FLOAT(J)*ARR(5)))-AMEAN)**2)*FLOAT(JSIZE(J))
          GDJFF=GDJFF+SDJFF
410        CONTINUE
          SDEV=SQRT(GDJFF/(ARR(10)-1.))

```

```
RDEV=10**SDEV
ARR(42)=RDEV
WRJTE(1,240) (ARR(K),K=1,50)
240  FORMAT(3A6,7F8.0,3(/,10F8.0),/,10F8.2)
WRJTE(1,241) ISIZE
241  FORMAT(10J4)
STOP
END
```


APPENDIX B

Samples of Computer-generated Data Sheets from the
Rapid Scan Automated Pyrite Analysis

THE PENNSYLVANIA STATE UNIVERSITY COAL RESEARCH SECTION
 ** RAPID SCAN AUTOMATED PYRITE ANALYSIS **

14-AUG-78

PSOC- 634.
 20X25# A

* RUN PARAMETERS *

PHOTOMULTIPLIER SPOT SIZE IN MICRONS: 2.
 TOTAL NUMBER OF POINTS SAMPLED: 800000.
 TOTAL LENGTH OF SCAN IN MILLIMETERS: 1600.002
 STAGE DRIVE SPEED IN MICRONS/SECOND: 800.
 SIZE OF DISCONTINUITY TO BE BRIDGED IN MICRONS: 20.

* DATA SUMMARY *

NUMBER OF PYRITE PARTICLES ENCOUNTERED: 376.
 VOLUME PERCENT PYRITE IN SAMPLE: 1.30
 SMALLEST PYRITE CHORD LENGTH IN MICRONS: 2.
 LARGEST PYRITE CHORD LENGTH IN MICRONS: 506.
 TOTAL PYRITE CHORD LENGTH IN MICRONS: 15438.

* DECILE STATISTICS *

CHORD SIZE IN MICRONS AT X% OF TOTAL PYRITE CHORD LENGTH:

10%	20%	30%	40%	50%	60%	70%	80%	90%	100%
18.	32.	50.	66.	88.	132.	258.	334.	406.	506.

PERCENTAGE OF CHORDS BELOW X MICRONS IN LENGTH:

10	20	30	40	50	100	200	500	1000	2000
35.%	57.%	66.%	74.%	79.%	91.%	96.%	100.%	100.%	100.%

* SUMMARY STATISTICS FROM LOG10-TRANSFORMED DATA *

MEAN CHORD LENGTH IN MICRONS: 18.40
 STANDARD DEVIATION IN MICRONS: 3.43
 CHI SQUARE VALUE: NOT COMPUTED
 B1*(1/2): NOT COMPUTED
 B2= NOT COMPUTED

14-AUG-78

PSOC- 634.
20X25# A

PYRITE CHORD SIZE DISTRIBUTION

CHORD SIZE (UM)	CUM %	FREQUENCY							
		5	10	15	20	25	30	35	
		V	V	V	V	V	V	V	
2	6.91	*****							
4	14.10	*****							
6	22.87	*****							
8	29.26	*****							
10	35.37	*****							
12	41.49	*****							
14	44.41	*****							
16	48.94	*****							
18	53.19	*****							
20	57.18	*****							
22	59.84	*****							
24	62.77	*****							
26	64.63	*****							
28	64.89	*							
30	66.49	*****							
32	68.62	*****							
34	71.54	*****							
36	72.61	****							
38	73.40	***							
40	74.47	****							
42	75.27	***							
44	76.33	****							
46	77.39	****							
48	77.66	*							
50	78.99	*****							
54	79.79	***							
56	80.59	***							
58	80.85	*							
60	81.91	****							
62	83.78	*****							
64	84.84	****							
66	85.37	**							
68	85.64	*							
70	86.44	***							
72	86.97	**							
74	87.23	*							
76	87.77	**							
78	88.30	**							
80	88.83	**							
82	100.00	*****							42