COMPREHENSIVE INVESTIGATION OF THE LIBERATION CHARACTERISTICS OF
PYRITE AND OTHER ASH-FORMING MINERALS FROM COAL

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FINAL REPORT

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ABSTRACT

The objective of this project is the development of methods for the measurement, prediction and modeling of the liberation characteristics of mineral matter and pyrite from coal, and to implement these findings in viable computer-simulation systems for coal cleaning plants. The central thrust of the project was the development of a convenient, accurate and experimentally verifiable method for the efficient measurement of the liberation spectrum of pyrite and other ash-forming minerals in coal. An image analysis method was developed that satisfied all these requirements and which was shown to identify these components in samples of coal particles and also whole coal. Images were generated by back scattered electron (BSE) detection in a scanning electron microscope. The intensity of the BSE signal was shown to be proportional to the mass weighted average atomic number of the phase so that individual mineral phase could be easily and precisely located in the images. The accuracy of the method was confirmed by comparing coal densities calculated using the measured phase volume fractions by image analysis with densities measured by helium pycnometry. The image analysis method was shown to be as accurate as measurements made by float-sink analysis using dense liquids.

A complete image analysis procedure was developed for handling both particulate and whole coal. The method included phase and particle-edge delineation artefact removal phase area measurement and linear intercept and linear grade distribution determination. Although operator supervision of image collection was required, all subsequent analysis was successfully automated.

Dense-liquid techniques were used to produce fractionated samples which were analysed using the image analysis method. These fractionated parent samples were subjected to controlled comminution and the image-analysis was used to measure the liberation spectrum in each of 7 progeny size fractions. The results showed clearly the pattern of liberation of both ash minerals and pyrite as particle size decreases. The matrix of cross transfer coefficients for that defines a simple Andrews-Mika diagram for the coal was established.

A model for the prediction of the liberation of mineral matter and pyrite from coal after comminution was developed based on a linear stochastic model for the description of the mineralogical texture and the random fracture pattern associated with the comminution process.

Stereological correction of the distribution of linear grades was required for both the measurement and prediction of the true distribution of volumetric grades in the particle population. A previously developed stereological correction method was successfully adapted to the three phase system that was studied.

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EXECUTIVE SUMMARY

The objectives of the project were:

1. Develop laboratory techniques for the quantitative measurement of the liberation spectra in samples of coal particles in the size range $38\mu\text{m}$ - 1 mm. Three dimensional spectra were required to account for the mineral matter and pyrite separately. A method based on quantitative image analysis was successfully developed.
2. Establish, experimentally, the liberation pattern for a typical U.S. coal and to develop an appropriate matrix form of the Andrews-Mika diagram so that it may be determined easily and quickly for coals of different origin and different type.
3. Establish an effective and reliable simulation technique so that liberation of both pyritic sulfur and ash during comminution operations can be modeled and the operation of coal preparation facilities simulated. These models will be incorporated into a computer simulation system for coal cleaning plants.

Each of these objectives was achieved.

The main significance of these results can be summarized as:

- a. The liberation spectra of coal in the particulate state can be accurately measured by image analysis. This technique will eventually supersede conventional dense-liquid laboratory methods entirely for coal in the sub millimeter size range. Potential savings in costs are large.
- b. It is possible to calculate the liberation of ash and pyrite from coal during comminution using data that can be easily determined by image analysis. Simulation of both batch and continuous comminution operations is possible.

One PhD thesis was produced and successfully defended based on the work of this project. Four papers have so far been published on the work. Several more are planned or are in preparation.

1 INTRODUCTION

Physical separation of pyrite and other ash-forming minerals from coal is possible only if these mineral phases can be liberated. Liberation is usually achieved by comminution. In general, the finer an ore is ground, the greater the degree of liberation that is achieved. However coal does not follow this simple pattern and it shows some unusual liberation effects. In addition the liberation characteristics of the separate phases in coal have never been quantified so it has not been possible to make quantitative calculations of the disposition of the ash minerals in coal after comminution. Liberation analysis has developed into a routine analytical method in mineral processing during the past decade and it is becoming steadily more widely used as a powerful diagnostic tool to assist in improving recoveries and grades of mineral concentrates. This study was undertaken to investigate comprehensively the liberation characteristics of a typical East Coast coal and to build up a methodology that will permit routine measurement of the liberation characteristics of coal and also provide a predictive model for calculating the liberation spectrum of a particular coal using easily implemented methods that do not rely on the use of dense liquids.

The method developed in this study is based on the use of image analysis techniques which have been well developed for more conventional mineral systems. This method was shown to be ideal for the measurement of liberation in coal smaller than 1mm. This method was found to be sufficiently efficient in the laboratory to analyze in detail the products obtained from the comminution of samples of coal and the liberation process could be observed in detail. The image analysis method was carefully checked to assess its accuracy and it was shown that the method is capable of providing predictions of coal densities almost as accurately as these can be measured by helium pycnometry and therefore the image analysis method rivals the dense-liquid method for accuracy and provides important additional information in the form of separate accounting for both pyrite and all other ash-forming minerals.

Mineral liberation is the mutual separation of a mineral phase from host ores by size reduction. The extent of mineral liberation can be evaluated by the degree of liberation or by the liberation spectrum. The degree of liberation of a particular mineral or phase is the percentage of that mineral or phase occurring as free particles in relation to the total of that mineral occurring in both the free and locked forms. The liberation spectrum is the distribution of ore by composition across the entire particle population. Obviously, the liberation spectrum gives a more complete description of mineral liberation than the degree of liberation or any other index of liberation. If the liberation spectrum is available, the degree of liberation can be calculated in a straight forward manner.

Measuring liberation is not an easy task. Traditionally, the liberation spectrum, which in coal

processing terminology is called the washability, is measured only for some special binary ore systems by the float-and-sink method. In those binary ore systems, the two phases must have sufficient density differences and their density should be in some manageable range. Among those ores, coal is the most favorable to the float-and-sink method. However, the float-and-sink experiment has several significant disadvantages. First of all, such an experiment needs a well-equipped laboratory and the experiment is time-consuming and tedious. Secondly, toxic organic liquids are used in such an experiment. They are unpleasant to handle. Third, this experiment is only available for some special binary ores, i.e. the two phases of the ore have to be significantly different in density and their densities should be in manageable ranges. If the densities of the two phases of interest are close to each other, it may be difficult to separate them by heavy liquid separation. If the concerned phases are too light or too heavy, there may be no appropriate liquids which could be found for the experiment. Fourth, it may not be possible to convert washability curves into liberation spectra. Washability curves only show the distribution of apparent density by ore mass. In order to transform washability into liberation spectrum, the relationship between apparent density and true density of the ore has to be found and the distribution of density at each pure phase has to be measured. In practice, because any binary ore systems only approximately exist, it is not easy to do the density transformation and to reach any of the pure phases. Finally, the float-and-sink experiment can be used only for binary ore systems. Actual ore systems, in most cases, are multi-phase. If more than two phases in ores are considered, the float-and-sink experiment yields only partial information. In particular we are interested specifically in the pyrite and ash as separate phases in coal so that it must be regarded as a ternary system.

Scanning electron microscopy (SEM) coupled with automated image analysis (AIA) has emerged as an important method for the measurement of liberation spectra (King and Schneider, 1993). However, the application of the SEM-AIA technique to the analysis of coal has some specific difficulties that must be addressed. Firstly, images taken from sectioned samples must be of good quality in order to acquire reliable measured results. Images of good quality mean that each of the phases of interest must be identifiable and must be precisely discriminated in the image and each must be clearly distinguished from the background mounting medium. Secondly, the SEM-AIA technique can provide only one-dimensional or two-dimensional information. Reliable stereological correction procedures are essential to use image analysis successfully.

This report provides a methodology for measuring liberation spectra for coal with MMIA (Metallurgical and mineralogical Image Analysis), a software for mineral liberation measurements developed at Utah Comminution Center. Linear liberation spectra for coal samples were measured first. Measured data were then transformed into volumetric grade distributions using the kernel function generated from PARGEN simulation program (Lin et al., 1988). Finally, the measured

results for coal Pittsburgh #8 1997 are discussed.

2 PREPARATION OF QUALITY IMAGES

Table 1 SEM parameter setting for imaging coal samples

Particle size μm	Accelerating voltage kV	Magnification	Working distance mm	Image resolution $\mu\text{m}/\text{pixel}$
1000×710	30	20X	44	
710×500	30	30X	29.3	
500×355	30	50X	17.6	
355×250	30	50X	17.6	
250×180	30	70X	14	
180×106	30	100X	14	
106×38	30	200X	14	

In this research, two samples of raw coal from the Pittsburgh number 8 seam were chosen to be tested because of its relatively high sulphur content and its high ratio of pyrite sulphur to organic sulphur. The samples were named as Pittsburgh #8 1994 and Pittsburgh #8 1997.

The coal sample was first crushed down to below 1000 μm with a roll crusher. The texture of the coal used was sufficiently fine to ensure that very little liberation of ash or pyrite occurs at sizes larger than 1000 μm . Particles below 1000 μm are appropriate to SEM-AIA processing.

Regular mounting media are carbon-rich materials, whose average atomic number is almost the same as that of pure coal. In this research, SEM equipped with both back-scattered and secondary electron detectors was used under the consideration of both cost and quality for the generation of images. In back-scattered SEM images, the grey level of each phase in an image is directly related to its average atomic number. If the medium and pure coal phase have similar average atomic numbers, they will have similar grey levels. As a result, it is difficult to distinguish between epoxy medium and pure coal phase. In order to increment the contrast between epoxy medium and pure coal phase, the differences between their average atomic numbers have to be created.

Figure 1 Typical image of coal particles from the 1.25x0 g/cc density fraction. The coal phase is dark grey, ash minerals are light grey and pyrite is white.

In this study, chemically pure iodoform was dissolved into regular Buehler epoxy resin to increase its average atomic number. Iodine has atomic number 53 which is almost ten times that of carbon. The more iodoform in the epoxy resin, the higher the background grey level in back scattered SEM images. The limiting content of iodoform in epoxy resin is determined by the solubility of iodoform in the epoxy resin. If iodoform cannot dissolve into epoxy resin, iodoform crystals will become inhomogeneous impurities in the plastic samples, which are artifacts and will certainly worsen image quality. Iodoform content should be appropriate to have background grey level somewhere between pure coal and the ash-forming minerals. Preliminary experiments demonstrated that approximately 13% or so by weight of iodoform in epoxy resin is suitable. When dissolving iodoform into epoxy resin, proper heating and stirring are necessary to speed up the dissolution. Sufficient stand time should be left before the iodized epoxy resin is to be used so as to completely get rid of undissolved iodoform crystals.

When mounting coal particles, bubbles might be introduced. Bubbles are another factor that degrade

Figure 2 Typical image of coal particles from 1.31x1.29 g/cc density fraction. Coal is dark grey, ash minerals are light grey and pyrite is white.

image quality. They might appear as a separate phase in SEM images. This problem has been well solved by mounting samples under a vacuum. In addition, MMIA provides a function to delete bubbles from images using the typical geometrical shape of bubbles in the image.

Figure 3 typical image of coal particles from the 1.60x1.50 g/cc density fraction. Coal is dark grey, ash minerals are light grey and pyrite is white.

Mounted samples should be well ground and polished to get good images. After many trials, five grinding and polishing stages have been adopted to produce good sectioned sample surface. Samples are firstly ground on 320 grit and 600 grit silicon carbide grinding paper with polishing oil. Subsequently, they are polished on polishing pads with 9 μm , 3 μm and 1 μm oil-based diamond suspensions sequentially.

After quality sectioned samples are prepared, image quality is determined by the imaging process. In a good image, pyrite should be saturated with the highest grey level up to 255, pure coal should be imbedded with the lowest grey level, and pure coal, background, ash-forming minerals and pyrite should separate each other very clearly. Therefore it is critical to set optimal SEM parameters.

Through preliminary experiment, standard settings for some SEM parameters were determined for different sizes of coal particles as shown in Table 1. Good separation of all phases of interest in

images can be estimated from both waveforms and image histograms. After some parameters are set according to Table 1, remaining parameters to be adjusted are contrast and brightness. Optimal contrast and brightness must be set to make full use of the available grey level range of the SEM.

Figures 1 to 4 show some typical images corresponding to coal samples from each density fraction. Coal particles are all from the 1000 μm and 710 μm fraction. In these images, pyrite is white, other ash-forming minerals are light grey, pure coal is dark grey and background (epoxy resin) is black. These grey levels have been chosen here to illustrate the features of the images in the comparatively low resolution of laser printers which have poor ability to represent subtle variations in grey level.

Figure 4 Typical image of coal particles from the +1.60 g/cc density fraction. Coal is dark grey appearing mostly as small inclusions in the ash which is the dominant light grey phase. Pyrite is white.

The images were acquired at high resolution and are accurately stored in 256 grey levels. The reproductions possible in a printed report do not do justice to the wealth of information that is available visually on a high resolution monitor screen. In particular it should be noted that grey levels

have been electronically switched to create printed images that have appropriate visual cues. In reality of course the measured grey level of the iodized epoxy is intermediate between those of the coal and the ash minerals as is explained in the next section. Approximately 2500 images were generated during the course of this investigation and these are each 1MB in size so it is impossible to include them in this report. All the images are indexed and archived and they constitute a valuable resource for any researcher who is interested in the image analysis of coal. These images can be supplied to interested persons on application to the Principal Investigator.

It is clear from these images that the dominant phases of interest are clearly identified. The trend in the character of the images as the density of the particles increases is striking and consistent. The lowest density fraction contains very little ash-forming minerals and virtually no pyrite. As the density of the samples increase the images reveal a steady increase in the amount of ash and pyrite. The ash phase dominates all the images in the +1.60 g/cc fraction. The geometrical characteristics of the separate phases are also well represented and identified in the images although this aspect of the image analysis requires further detailed attention. This is described later in this report.

3 IMAGE ANALYSIS OF COAL

The foundation of the work undertaken during this project is based on the observation that image analysis of properly prepared samples reveals that coal can be regarded as a mineralogical material

Figure 5 Measured grey level histogram of the +1.60 g/cc fraction. Histogram averaged over 40 fields of view.

consisting of three more or less distinct phases. This is clearly evident from the images shown in

Figures 1 to 4. The different phases in the coal produce sufficiently different BSE image intensities to reveal the texture of the coal with respect to these phases. The separation of the phases is revealed quantitatively in the grey-level histogram shown in Figure 5. The peaks corresponding to coal (grey level 6), iodized epoxy (grey level 27), ash minerals (grey level 78), and pyrite (grey level 222) are distinctively separated in the histogram, although there is significant overlap of the tails of neighboring peaks by the primary peaks. The distinct peaks in the histogram confirm that quantitative analysis of coal as a three-component mineral is distinctly possible, albeit with careful attention to the image analysis procedures that are used.

The BSE signal intensity from any point on the section through the solid is proportional to the average atomic number of the material at that point in the image. This is demonstrated by the data in Figure 8 which shows the measured BSE intensity against the average atomic number of a number of widely different minerals obtained with both Robinson and solid-state BSE detectors.

Figure 6 Relationship between BSE detector intensity and the average atomic number.

The slope of the lines in Figure 6 depends on the contrast setting of the SEM. The higher the contrast the steeper the line, but the narrower the total range of atomic numbers that can be covered in a single image. The BSE intensities that are plotted are determined from the positions of the peaks in the image histogram such as that shown in Figure 6. The average atomic number of pyrite is 20.57 so that the uppermost point on the coal line in Figure 6 is fixed. The average atomic number of coal

is approximately equal to that of the epoxy without iodine since these materials have very similar BSE responses. This can be used to fix a second point on the coal line in Figure 6. Once the grey levels for two minerals of known elemental composition have been determined, grey levels in the image can be converted to average atomic number using the formula

$$\bar{N} = \frac{gl - gl_0}{gl_1 - gl_0}(N_1 - N_0) \quad (1)$$

where

$\bar{N}$ = average atomic number of phase having grey level gl .

gl = grey level

gl_0 = grey level for phase 0

gl_1 = grey level for phase 1

N_0 = average atomic number of phase 0

N_1 = average atomic number of phase 1.

Using the data from Figure 6

$$\bar{N} = \frac{gl - 6}{222 - 6}(20.65 - 5.4) \quad (2)$$

The histograms can then be plotted as intensity spectra with average atomic number as the independent variable. Three typical spectra for the coal sample are plotted in Figures 7 to 9.

Figure 7 Data from Figure 5 plotted as an intensity spectrum for the +1.60 g/cc fraction, 1000x710 μm .

Figure 7 shows the spectrum for the densest fraction +1.60g/cc. The spectrum shows four distinct peaks: the coal at average atomic number 5.11, the iodized epoxy at average atomic number 6.62, a fairly broad peak centered at average atomic number 10.3 and a weak broad peak at average atomic number 20.6 corresponding to pyrite.

The major minerals that form ash in Pittsburgh#8 coal have been determined by Straszheim, Yousling and Markuszewski (1986), These are shown in Table 3 together with their average atomic numbers calculated from the elemental composition of the minerals.

Table 3 Ash-forming minerals identified in Pittsburgh#8 coal. (Straszheim *et al.* 1986)

Mineral	Chemical formula	Average atomic number	Density kg/m ³
Pyrite	FeS ₂	20.65	5000
Marcasite	FeS ₂	20.65	4900
Quartz	SiO ₂	10.57	2670
Kaolinite	Al ₂ Si ₂ O ₅ (OH) ₄	10.24	2600
Illite	KAl ₂ (AlSi ₃ O ₁₀)(OH) ₂	11.33	
Calcite	CaCO ₃	12.56	2710
Siderite	FeCO ₃	16.47	3850
Gypsum	CaSO ₄	12.12	2320
Dolomite	Mg,Ca(CO ₃) ₂	10.87	2850
Rutile	TiO ₂	16.39	4020
Alumina	Al ₂ O ₃	10.65	4000

Figure 8 Typical image of vein form of pyrite in a coal particle.

The densest fraction of the coal consists largely of the minerals quartz (10.57) and kaolinite(10.24) with a small quantity of pyrite.

Figure 9 Typical lense form of pyrite in a coal particle.

Sulfur occurs in coal in three forms. It is present in organic combination as part of the coal phase, as sulfate and as pyrite or marcasite. The image analysis technique that was developed here cannot identify organic sulfur. Sulfates are not usually present in large quantities but even small grains of CaSO_4 can be detected in the BSE signal as discussed below. The pyrite and marcasite have different crystalline forms and they have slightly different BSE responses. In this work they are considered together as pyrite. Macroscopic pyrite occurs in pyrite predominantly in four forms: veins, lenses, balls and pyritized plant tissue (Leonard, 19). The first three of these forms were observed in this study and typical images are shown in Figures 8 - 10. These images were extracted from the larger images of the +1.60 g/cc density fraction.

Figure 10 Typical nodule form of pyrite in a coal particle.

The pyrite peak is quite broad because the pyrite is finely disseminated with only a comparatively few large grains. Some typical images of individual coal particles containing pyrite are shown in Figures 8 - 10. These illustrate typical crystal forms of pyrite that are commonly found in coal. The BSE intensity from pixels on the edge of a grain is smaller than that from a pixel in the interior of a grain because of the finite spot size in the SEM. This leads to a spread out tail on the low intensity side of the pyrite peak.

Figure 11 BSE intensity spectrum of a single field of view of the +1.60g/cc fraction. Particle size is 1000x710 μm

Other minerals are present in small quantities and were detected on one or two specific fields of view as single isolated grains. The spectrum of a single field of view is shown in Figure 11. This field contained a single grain of gypsum and this shows up clearly in the spectrum. Fields of view that contain fairly large pyrite grains generate a well-defined pyrite peak as shown in Figure 12.

Figure 12 Spectrum from a single field of view of the +1.60 g/cc showing a well-defined pyrite peak.

The mineral content of the less dense fractions differs significantly from that in the +1.60 g/cc fraction. This is obvious from the spectrum shown in Figure 13 which shows the BSE intensity

Figure 13 BSE intensity spectrum for the 1.60x1.50 g/cc fraction. Histogram averaged over 40 fields of view.

spectrum for the 1.60x1.50 g/cc fraction. The ash minerals are finely disseminated and are associated with the coal. This shows up as a broad tail on the coal peak that cuts across the background peak. The silica and pyrite peaks are small but clearly discernible.

Figure 14 BSE intensity spectrum of the - 1.25 g/cc fraction. Histogram averaged over 40 fields of view.

The spectrum of the lightest fraction shown in Figure 14 reveals that both the background and the coal peaks are sharp and clearly defined. In the absence of any significant quantity of ash minerals, the coal peak does not show any tail. This confirms that the coal components can be well discriminated in the images.

Image analysis with MMIA in this research was totally automated after appropriately setting processing parameters so that a large number of images could be processed within a reasonable period.

To increase the accuracy of phase identification, most of the images collected during this study, including those shown in Figures 1- 4, were obtained using a high-contrast mode of operation. The contrast level was increased to stretch the histogram until all pixels in the coal phase were always recorded at grey value zero and pyrite was saturated at 255. This gave maximum peak separation in the histogram and helped to allocate pixels to the correct phase.

4 CALIBRATION OF THE IMAGE ANALYSIS METHOD

The image analysis method was developed for the automatic detection and quantitative measurement of the three component "phases" of any coal sample. The limitations that are inherent in the image analysis method preclude this technique as an absolute analytical method. These limitations include the finite resolution of the images and the natural variation in the composition of each of the three "phases." The intensity of the back scattered electrons signal from the surface is proportional to the

average atomic number of the material under the electron beam once the operating parameters (accelerating voltage, spot size, working distance, brightness and contrast) have been set. Small variations in the composition on the surface can lead to erroneous signal levels particularly in the neighborhood of a phase boundary. The histogram of grey levels in Figure 5 shows that although the peaks for each "phase" can be well separated from each other, and the peak for the epoxy background can be partitioned between the coal and ash peaks, some overlap is inevitable. This leads to misidentification of the phase for some pixels in the image. Consequently it is necessary to calibrate the image analysis method against some independent analytical technique.

Two independent methods were considered: proximate analysis and accurate measurement of particle density by helium pycnometry. Of these the latter is the preferred method since we are ultimately interested in predicting the density spectrum of comminuted coal particles which in turn determines the performance of the coal in coal washing operations.

4.1 Proximate Analysis

A 710-1000 micron size fraction of a sample of Pittsburgh #8 1994 coal was carefully fractionated using precise dense-liquid separation techniques. The sample was fractionated at 1.25, 1.27, 1.29, 1.31, 1.33, 1.35, 1.37 1.39, 1.50 and 1.60 grams per cc.

Table 2 Proximate analysis of density fractions of the Pittsburgh#8-1994 sample

Density range g/cc	Midpoint density g/cc	Weight %	Moisture %	Ash %	Fixed Carbon %	Volatiles %	Sulfur %	Density measured by helium pycnometry	w ₁	w ₂	w ₃	Calculated density g/cc
<1.25		2.02	1.81	1.35	57.63	39.21	1.538	1.286	0.9233	0.0029	0.0738	1.325
1.25 - 1.27	1.26	12.8	1.84	1.90	57.91	38.35	1.625	1.296	0.9172	0.0047	0.0781	1.330
1.27 - 0.29	1.28	17.7	1.80	3.11	57.40	37.69	1.808	1.310	0.9040	0.0085	0.0875	1.341
1.29 - 1.39	1.34	52.3	0.30	7.49	55.84	36.37	2.472	1.347	0.8560	0.0223	0.1217	1.382
1.39 - 1.50	1.445	5.57	0.36	16.24	50.51	32.91	4.787	1.472	0.7551	0.0686	0.1764	1.485
1.50 - 1.60	1.55	1.61	0.30	24.50	46.21	29.00	5.386	1.568	0.6678	0.0823	0.2499	1.575
> 1.60		7.99	0.26	55.24	25.04	19.46	8.860	2.077	0.3367	0.1568	0.5065	2.066
Composite		100	0.49	12.28	52.04	35.00	3.182	1.400	0.8035	0.0371	0.1594	1.430
Reconstituted composite			0.79	10.45	53.50	35.25	2.91	1.380	0.8235	0.0315	0.1450	1.411
0.29 - 1.31	1.30	55.72	0.49	5.65	56.77	37.09	1.98		0.8515	0.0132	0.1353	1.383
1.31 - 1.33	1.32	19.46	0.37	7.89	55.64	36.10	2.62		0.8266	0.0259	0.1476	1.407
1.33 - 0.35	1.34	11.68	0.57	9.33	54.61	35.46	3.01		0.8106	0.0337	0.1558	1.423
1.35 - 1.37	1.36	9.25	0.54	11.10	53.40	34.96	3.52		0.7908	0.0437	0.1654	1.444
1.37 - 1.39	1.38	3.89	0.25	13.78	52.04	33.93	4.21		0.7612	0.0576	0.1813	1.475
Reconstituted 1.29 - 1.39			0.47	7.34	55.80	36.39	2.45					

the proximate analysis was determined for each of the density fractions, together with measurement of sulfur content and average particle density. The results are shown in Table 2. The composite samples in the 1.29 - 1.39 g/cc fraction and the total sample composite were analyzed directly and the reconstituted values were calculated to assure the quality of the analysis. The results are shown in Table 2 and the reconstituted values are in good agreement with those measured directly.

Figure 15 Correlation between sample density measured by helium pycnometry and the mid point density of the dense liquid fractions.

The average particle densities are particularly important in establishing the precision of the fractionation procedure because it is ultimately the density of the particles that will determine their behavior in a coal washing operation. The average solid densities of the air-dried coal were measured using helium pycnometry. The results for samples of Pittsburgh #8 1994 and 1997 are shown in Figure 15. The density measured by helium pycnometry is, in all cases, slightly higher than the mid-point of the fractionation interval and this is consistent with the observation that helium pycnometry registers a higher solid density because much smaller pores are accessible to the helium and are not accessible to liquids because of capillary pressure. The strong linear relationship between true density from pycnometry and the dense-liquid midpoint density is clearly evident. The least squares best linear relationships is $\rho_{pyc} = 0.1671 + 0.8956 \times \text{midpoint density}$.

In order to calibrate the image analysis results, it was necessary to estimate the quantities of the three phases from the proximate analyses. In order to do so, a simple model for the three-component system was constructed based on the following assumptions: a constant ratio, γ , relates mineral matter in the coal to measured ash; and the coal contains a constant fraction, s , of organic sulfur and

constant fraction, c , of inherent mineral matter. In addition, all pyrite is assumed to convert to Fe_2O_3 on ashing, and pyrite contains 53.3 percent of sulfur. A simple mass balance allows the calculation of the fraction of pyrite and mineral matter-free coal in any sample in which ash and sulfur are determined. The model is described by:

$$\begin{aligned} A &= 0.664 w_2 + \gamma(cw_1 + w_3) \\ S &= 0.533 w_2 + sw_1 \\ w_1 + w_2 + w_3 &= 1 \end{aligned} \quad (3)$$

where A and S are measured ash and sulfur fractions respectively and w_1 , w_2 and w_3 are the calculated mass fractions of coal, pyrite and mineral matter. The efficacy of this simple model can be tested by comparing the calculated density of the separated fractions to the mid-point of the separated fraction using

$$\frac{1}{\rho} = \frac{w_1}{\rho_c} + \frac{w_2}{\rho_p} + \frac{w_3}{\rho_m} \quad (4)$$

where ρ_c , ρ_p and ρ_m are the densities of the coal, the pyrite and the mineral matter respectively. The data using $\rho_c = 1.27$, $\rho_p = 5.0$, $\rho_m = 2.7$, $\gamma = 0.926$, $s = 0.015$ and $c = 0.0135$ are shown in Table 2. The effectiveness of this simple model can be established by noting that the specific volume of the sample is related to these quantities by Equation 2. The predicted densities from equation 2 can be compared to the densities measured by helium pycnometry in Table 2. The agreements are satisfactory in spite of the model being based on a number of assumed parameter values. The calculated densities are about 0.03 g/cc higher than the measured values. These uncertainties make the use of proximate analysis uncertain as a method to calibrate the image analysis procedure. A more direct method was accordingly chosen as described in the next section.

4.2 Density Analysis

The density of a composite coal particle can be related to the density of the three-component phases by

$$\rho = v_1\rho_c + v_2\rho_p + v_3\rho_m \quad (5)$$

where v_1 , v_2 , and v_3 are the volume fractions of each phase. This equation provides a more direct calibration method that is more suited to the needs of this project.

The measurement of the volume fraction of individual phases is the simplest quantitative measurement to make using image analysis. The volume fraction of a phase is equal to the area fraction of that phase that is revealed in a random section through the particle population. In a digital image, the area fraction is just the number of pixels that are allocated to the phase in a sufficiently large number of sections to ensure that the pixel counts are representative of the phases. The only difficulty lies in allocating the appropriate phase to each and every pixel in the images.

The grey-value histogram in Figure 5 can be resolved into the overlapping single peaks. A rough estimate of the area fraction of each phase is just the area under each respective peak. However this cannot be regarded as a satisfactory way to measure area fraction unless the histogram peaks are cleanly separated.. Pixels that have grey values that fall in the region of overlap of two peaks such as those that characterize ash and iodized epoxy in Figure 10. Additional information from the image must be used to allocate pixels to phases in the image.

The histogram gives no information about the texture of the images, and consequently it is necessary to identify the phase at each pixel in the image. Because of the overlap of the intensity peaks it is not possible to identify each pixel unambiguously, and additional information must be incorporated into the identification algorithm. This is done using delineation filtering of the image. Delineation filtering is based on the following observations. In any region of the image where the measured grey value is fairly uniform, the phase identify of the pixel is determined by the grey value alone. These regions correspond to relatively large areas of a single phase and therefore are unambiguously identified by grey value only. In regions where the measured grey value varies rapidly from pixel to pixel, it is necessary to consider both the grey value of the pixel and the grey values of all the neighboring pixels to allocate a phase to the pixel in question. This is done by considering the 5x5 pixel environment of each pixel. The grey value of the pixel is adjusted to either the minimum or maximum neighborhood value depending on whether the original grey value of the pixel is below or above a test value that lies between the neighborhood minimum or maximum. Usually the test value is the median grey value for the neighborhood, but other choices are possible depending on the mineralogical texture. In this work we used

$$g_{test} = 0.5(g_{min} + g_{max}) \quad \text{when } g_{max} < 2.50 \quad (6)$$

$$g_{test} = 0.99g_{max} \quad \text{when } g_{max} \geq 2.50 \quad (7)$$

Equation (7) was used to prevent excessive incorrect allocation of ash pixels to the pyrite phase because of the stretching of the central region of the histogram to increase the separation between the BSE intensity of the coal phase and that of the iodized epoxy and the separation between the iodized epoxy and the ash. Delineation filtering has the effect of shifting grey values from the regions of the histogram between the phase peaks into the peak regions and so effectively allowing cleaner discrimination of the phases.

It is easy to see that a delineation filter will handle the problem of correctly allocating an isolated pixel having a grey value within the iodized epoxy peak in the histogram but which is detected within the coal or ash phase. The delineation filter will allocate the pixel correctly to the neighboring phase in almost every case. In other words pixels must not only have the correct grey value but they must have some association in the neighborhood with the phase to which they are allocated.

Delineation filtering also eliminates halo formation which is a problem that is common when phase identification is attempted on a digital image. Because of the finite size of the electron beam spot

Figure 17 Calibration of the image analysis method using density prediction..

size and the comparatively coarse resolution of each pixel, pixels having intermediate grey values will always exist between a bright and darker phase. If a third phase exists at grey values between these two phases, the intermediate grey values will often be incorrectly allocated to the third phase, which creates a distinct halo that surrounds the brighter phase.

MMIA has been specially adapted for processing coal samples. It can recognize phase boundary,

remove noise, and measure linear liberation spectra. Its major functions can be found in King and Schneider (1993). In this work the linear grade distributions were the primary concern.

After delineation filtering it is only necessary to run a grey level segmentation operation on the images to allocate all of the pixels in the image to the correct phases. The volume fraction of a phase is then calculated as the fraction of pixels in that phase as a fraction of the total number of pixels that are in particles in the image.

The reliability of the allocation algorithm can be judged from the measured area fractions of each of the three phases and using equation 5 to calculate the density of the sample. The measured area fractions for each phase in each of the parent samples are given in Table 3 together with the calculated sample densities from equation 5. The densities of the separate phases are $\rho_c = 1.28$, $\rho_m = 2.7$ and $\rho_p = 5.0$ g/cc. These values were then compared to those measured by helium pycnometry. The results are shown in Figure 16. The correspondence is good confirming that the image analysis procedure is capable of accurately identifying the phases and making good predictions of the particle densities.

Table 3 Comparison of sample density from helium pycnometry and image analysis.

Density range g/cc	Measured area fraction			Density calculated from measured area fraction using equation (3). g/cc	Measured density g/cc
	Coal	Ash	Pyrite		
<1.25	99.38	0.57	0.05	1.28	1.278
1.25 - 1.27	98.46	1.45	0.09	1.29	1.296
1.27 - 1.29	97.81	2.05	0.14	1.30	1.313
1.29 - 1.31	96.74	2.53	0.72	1.33	1.334
1.31 - 1.33	95.46	3.27	1.27	1.36	1.353
1.33 - 1.35	94.97	3.97	1.06	1.37	1.365
1.35 - 1.37	93.31	5.75	0.94	1.39	1.385
1.37 - 1.39	93.38	5.42	1.21	1.39	1.402
1.39 - 1.45	90.03	7.68	2.28	1.46	1.439
1.45 - 1.50	90.48	4.96	4.56	1.51	1.490
1.50 - 1.60	87.07	8.05	4.88	1.57	1.553
>1.60	11.14	80.43	8.43	2.73	2.513

After processing images, MMIA also output the values of average volumetric grades for all interested phases in a sample. Average volumetric grade is a reliable measured result and can be used directly. Average volumetric grades of phases for coal samples in different size ranges and

different density ranges can also be used to evaluate the liberation characteristics for coal. Table 3 gives the average volumetric grades of each phase in coal in different density ranges and different size ranges. It can be seen that in low density range, particles consist mainly of pure coal phase, in high density range, particles consist mainly of pyrite and other ash. In the middle density range, pure coal is still the major phase, but there is also significant amounts of pyrite and other ash-forming minerals in the samples. After breakage, particles with small size contain more pyrite and other ash-forming minerals and less pure coal phase than those with large size. The tendency for ash and pyrite to accumulate in smaller sizes during comminution is a commonly observed phenomenon and is caused by selective or preferential breakage of the harder but more brittle inorganic components. For high density samples, the particles with small size contain more pure coal and less other ash-forming matters than those with large size.

5 DISPOSITION OF ASH AND PYRITE AFTER COMMUNUTION.

5.1 Measured using dense-liquid fractionation.

The narrow 1.29-1.31 fraction of Pittsburgh#8-1994 generated as described in section 4.1 was crushed in the ultrasonic mill in the Comminution Center laboratory and progeny larger than 38

Table 4 Disposition of progeny from comminution of 1000×710 μm and 1.31×1.29 g/cc particles with respect to size and density as determined by dense-liquid analysis.

Size distribution		Density fractions											
Size μm	%	-1.25	-1.27 +1.25	-1.29 +1.27	-1.31 +1.29	-1.33 +1.31	-1.35 +1.33	-1.37 +1.35	-1.39 +1.37	-1.45 +1.39	-1.50 +1.45	+1.50	Total
710×500	37.3	0.36	2.35	14.6	37.1	11.39	13.20	3.98	6.33	7.23	2.89	0.54	100.00
500×355	31.6	0.43	2.77	14.5	30.3	10.87	14.07	4.48	6.82	10.45	4.05	1.28	100.00
355×250	13.9	0.49	3.88	12.6	27.7	8.74	13.59	4.37	7.28	13.59	5.34	2.43	100.00
250×180	6.3	1.06	4.26	10.6	25.5	9.57	13.83	5.32	5.32	14.89	6.38	3.19	100.00
180×106	6.1	1.11	3.33	7.8	22.2	6.67	13.33	5.56	7.78	20.00	6.67	5.56	100.00
106×38	4.8	1.41	5.63	7.0	12.7	7.04	14.08	8.45	8.45	22.54	5.63	7.04	100.00

microns were fractionated at 1.25, 1.27, 1.29, 1.31, 1.33, 1.35, 1.37, 1.39, 1.45, 1.50 and 1.60 g/cc. Each of these fractions was then screened at 500, 355, 251, 180 and 160 microns so that the complete disposition of the progeny particle population could be established. These data define what may be regarded as a coarse-grained experimental version of the Andrews-Mika diagram for parent particles at 710-1000 microns and 1.31×1.29 g/cc.

The data in Table 4 and Figure 17 show a typical liberation pattern. As the progeny get finer, the distribution becomes more dispersed with more particles appearing at the liberated ends of the distribution. This data also show clearly that the separate phases liberate very slowly as the progeny

size decreases. It is necessary to reduce the size considerably below $38\mu\text{m}$ before substantial liberation can be expected. The experimental procedure described above is extremely tedious and in addition it does not reveal how the separate pyrite and other ash-forming minerals dispose themselves among the progeny sizes as comminution proceeds. It can reveal only how the density of the progeny is distributed after comminution. A better method is obviously called for and an effective method based on image analysis was developed during this study. This method was used to investigate the distribution of pyrite and ash throughout the progeny population resulting from the comminution of each of the parent density fractions.

Figure 17 Disposition of progeny particles following comminution of $1000 \times 710 \mu\text{m}$, $1.31 \times 1.29 \text{ g/cc}$ parent particles

5.2 Measured using image analysis.

The efficiency of the image analysis technique made it possible to measure the distribution of both ash and pyrite separately in the progeny from the breakage of all the parent particles. The Pittsburgh#8-1997 sample was used for all experiments that are described in this section.

5.2.1 Distribution of pyrite and ash in the parent particles.

The parent particle for this series of experiments were the density fractions prepared from the $1000 \times 710 \mu\text{m}$ fraction of the Pittsburgh#8-1997 sample. A representative sample of each of these

fractions was mounted, sectioned and polished as described previously for image by scanning

Measured linear grade distribution
Parent particles 1.60x1.37 g/cc 1000x710 μm

Figure 18 Measured linear grade distribution for the 1.31x1.29 g/cc parent fraction.

electron microscopy. Typical images are shown in Figures 1 to 4. After careful delineation filtering, the separate phases were identified by grey level discrimination. After discrimination the average volume fraction of each of the three phases is measured as the area fraction of the particles that consists of each phase. The results for the parent particles are given in Table 3.

Parent particles 1.25x0 g/cc 1000x710 μm

Figure 19 Stereologically corrected phase distributions for the 1.25x0 g/cc fraction of the parent particles.

The distribution of the phases in the particles was measured by linear intercept analysis (King 1994). A series of linear intercepts are inscribed across the particles and the linear grade of each line is

Parent particles 1.27x1.25 g/cc 1000x710 μm

Figure 20 Stereologically corrected phase distributions for the 1.27x1.25 g/cc fraction of the parent particles.

determined by calculating the fraction of the intercept length that is in each phase. Each intercept line therefore has associated with it three numbers which represent the fraction of the line in each

Parent particles 1.29x1.27g/cc 1000x710 μm

Figure 21 Stereologically corrected distribution of phases in the 1.29x1.27 g/cc fraction of the parent particles.

phase. The fractions sum to unity so only two of the three numbers are independent. Although these intercept lines are required to be random in orientation and position, experience has shown that all

Parent particles 1.31x1.29g/cc 1000x710 μm

Figure 22 Stereologically corrected phase distributions for the 1.31x1.29 g/cc fraction of the parent particles.

the subsequent analysis is valid for parallel lines that are separated by 10 or more pixels. The use of parallel lines is convenient and has been shown to be equivalent to randomly drawn lines.

Parent particles 1.37x1.33 g/cc 1000x710 μm

Figure 23 Stereologically corrected phase distribution for the parent particles in the 1.37x1.33 g/cc density fraction..

Several thousand intercepts (between 2000 and 5000 depending on magnification) were collected from a typical sample. Usually about 10 intercepts are recovered from each particle section. The individual intercepts were then sorted according to the linear grades into 43 classes or "bins". These

Parent particles 1.60x1.37 g/cc 1000x710 μm

Figure 24 Stereologically corrected phase distribution for the 1.60x1.37 g/cc parent particles.

43 grade classes include a class for each pure phases (3 classes), 15 classes for the binary

Parent particles +1.60 g/cc 1000x710 μm

Figure 25 Stereologically corrected phase distribution in the +1.60 g/cc fraction of the parent particles.

combinations in 20% increments and 25 classes for ternary combinations in 20% increments. A

typical graphical representation of the distribution of linear grades is shown in Figure 18. The triangular bar chart is used for convenience. The pure phases are represented by the bars at the three vertices of the triangle while the binary combinations are represented by the bars along the outer edges of the triangle. The 25 ternary combinations are represented by the bars in the interior of the chart.

The linear grade distribution does not represent directly the true volumetric grade distribution among the three dimensional particles because of the stereological error that is associated with the analysis of the 2-dimensional sections through the particles that are observed to generate the original images. The stereological correction from linear to volumetric distributions is not a straight forward procedure and specialized methods are required. A useful and effective procedure was developed for this calculation and it is described in detail by King and Schneider, (1998). All linear grade distributions that were measured during this project were stereologically corrected in this way. The measured volumetric distributions for the parent particle fractions are shown in Figures 19 to 25.

The pattern of ash and pyrite distributions in the parent particles is readily apparent from these diagrams. The lightest fraction (1.25x0 g/cc) Consist almost entirely of particles that contain more than 80% coal and less than 20% combined for the other two phases. These particles are represented by the three tall bars in Figure 20. It is interesting that although about 25% of the particles are binary coal-ash mixtures with less than 20% ash, there are virtually no coal - pyrite binaries. In fact pyrite occurs only in particles that contain ash as well. The coal - pyrite edge of the chart is devoid of particles. As the density of the parent sample increases, the distribution moves steadily into the interior of the chart and away from the coal apex. This trend is easily observed in the sequence of Figures from 19 to 23. Almost all particles in the 1.60x1.37 fraction are ternary mixtures with very few binary or pure single-phase particles. The heaviest fraction (+1.60 g/cc) has particles concentrated along the ash - pyrite binary edge but with many particles distributed throughout the three-phase interior of the chart.

The importance of proper stereological correction is evident by comparing Figures 18 and 24. The stereological bias towards the liberated edges of the diagram obscures the true nature of the distribution and would preclude the accurate calculation of the density of the coal particles without proper correction.

5.2.2 Disposition of the phases after comminution.

The image analysis method made it possible to measure accurately the disposition of the three phase during comminution from each of the parent density fractions. This is precisely the information that

is needed to calculate how the ash and pyrite are liberated during comminution. This calculation method has been described by King and Schneider(1992, 1995, 1999). The method is based on the same principle as the well-known Andrews-Mika diagram for two-phase systems. A matrix of cross transfer coefficients from every possible parent particle to every possible attainable particle type must be determined. Obviously it is impossible to measure every possible transfer coefficient because there are so very many of them (approximately 2×10^6) and we attempt to discern the pattern from observations on the breakage of the carefully selected parent samples which span the full range of parent particle types. Even this restricted approach to the problem generated a considerable number of samples for analysis and it was only the relative efficiency and accuracy of the image analysis method that made it possible during this project. Results from only one parent sample are included in this report in the interests of space. The fraction was chosen to illustrate the principle.

The monodensity and monosize parent particles were broken using a continuous hammer mill and then the products were separated by wet and dry screening. In order to do some comparison, a batch ultrasonic mill and a continuous ultrasonic mill without vibration were also used to make several progeny samples. The particle size distribution and average density for all progeny samples were listed in Table 5 and Table 6. Some of the parent density fractions were ground in an ultrasonic mill which was developed in the Comminution Center for controlled comminution. The ultrasonic mill does not produce an excess of fines and the progeny size distribution is more or less uniform over the six size intervals that were analyzed. The size distributions of the progeny for each parent sample are given in Table 5.

Table 5 Measured size distributions of the progeny from the controlled comminution of seven parent samples.

Sieve size, μm	Parent-sample density, g/cc									
	-1.25	-1.27+1.25	-1.27+1.25*	-1.29+1.27	-1.31+1.29	-1.31+1.29**	-1.33+1.31	-1.37+1.33	-1.6+1.37	+1.6
-710+500	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00
-500+355	47.17	50.10	49.79	55.75	49.90	42.48	49.42	50.95	44.70	45.38
-355+250	23.91	26.30	26.96	28.93	26.65	19.67	26.97	27.50	24.09	24.28
-250+180	13.69	16.18	15.77	17.31	17.08	10.16	17.49	17.91	15.72	16.22
-180+106	8.26	10.75	9.68	11.4	12.04	6.16	12.53	13.01	11.64	12.3
-106+38	3.04	5.53	3.48	5.28	5.76	3.67	6.70	6.61	6.33	8.91
-38+0	0.87	1.77	0.65	2.01	2.06	0.86	2.62	2.56	2.45	6.26

* Broken by continuous ultrasonic mill without vibration

** Broken by batch ultrasonic mill

The densities of the progeny particles were accurately measure using helium pycnometry and the results are shown in Table 6 and Figure 30. The density of the progeny shows a distinct trend to increase as the size decreases. This is a typical pattern of behavior for coal that is often observed. The inorganic ash-forming minerals are more brittle than the coal and consequently break down to

the finer sizes. This phenomenon could also be caused by preferential breakage along mineral coal interfaces so that the finely disseminated mineral particles are preferentially liberated.

Table 6 Pycnometer Density of Progeny Samples

Sieve size μm	Average size μm	Parent sample density, g/cc									
		-1.25	-1.27+1.25	-1.27+1.25*	-1.29+1.27	-1.31+1.29	-1.31+1.29**	-1.33+1.31	-1.37+1.33	-1.6+1.37	+1.6
-710+500	595.8188	1.262	1.281	1.291	1.308	1.315	1.317	1.338	1.364	1.454	2.515
-500+355	421.3075	1.269	1.283	1.297	1.315	1.317	1.319	1.341	1.366	1.453	2.49
-355+250	297.9094	1.269	1.287	1.307	1.321	1.326	1.323	1.341	1.374	1.461	2.471
-250+180	212.1320	1.272	1.289	1.328	1.326	1.329	1.328	1.343	1.397	1.478	2.473
-180+106	138.1304	1.293	1.320	1.356	1.340	1.343	1.338	1.361	1.412	1.488	2.453
-106+38	63.4665	1.316	1.327	1.448	1.349	1.354	1.365	1.375	1.444	1.552	2.518
-38+0	29.1900	1.409	1.371	2.018	1.355	1.377	1.548	1.388	1.457	1.553	2.431
Composite		1.274	1.295	1.295	1.310	1.334	1.334	1.353	1.375	1.464	2.512

* Broken by continuous ultrasonic mill without vibration

** Broken by batch ultrasonic mill

Figure 28 Phase distributions in the 710x500 μm progeny fraction from the 1.33x1.31 g/cc parent.

Figure 29 Phase distribution in the 355x250 μm progeny fraction from the 1.33x1.31 g/cc parent.

The density of the progeny particles does not provide a detailed picture of the liberation process because it does not reveal how the separate phases liberate during the comminution operation. The grade distributions of the progeny particles were therefore measured using the image analysis method that was developed in this project. This generated a large number of grade distributions and so only a single set from the 1.33x1.31 g/cc density fraction is shown here. The other parent particles produced results that were qualitatively similar. The measured distributions are shown in Figures

28 to 31. The pattern of liberation of the three phases can be seen in Figures 28 to 31. There is a distinct migration of particles from the inner triangular area representing the three-phase particles to the outer edges of the triangle that represent the binary and single-phase particles. In order to use this information to predict the liberation of ash and pyrite during comminution, a simple quantitative

Figure 30 Phase distribution in the 180x106 μm progeny fraction from the 1.33x1.31 g/cc parent fraction.

Figure 31 Phase distribution in the 106x38 μm progeny fraction from the 1.33x1.31 g/cc parent fraction.

scheme has been developed in which the fractions of particles in the seven broad grade classes representing the three pure phases (A,B,C), the three possible binaries (AB,BC,AC) and the total ternary(ABC) are given in matrix form in the following tables. A represents the coal phase, B the ash and C the pyrite.

Table 7 Measured liberation pattern of 1.25x0 g/cc parent

	Parent	710x500	500x355	355x250	250x180	180x106	106x38
A	29.70	29.68	26.01	46.19	26.38	22.70	3.59
B	0.00	0.00	0.00	0.00	0.00	0.03	0.29
C	0.00	0.00	0.00	0.00	0.00	0.10	0.02
AB	27.16	27.44	30.14	21.29	29.26	20.37	40.91
AC	0.23	0.24	0.24	0.16	0.24	2.29	0.24
BC	0.00	0.00	0.00	0.00	0.00	0.00	0.03
ABC	42.91	42.64	43.61	32.36	44.13	54.51	54.92

Table 8 Measured liberation pattern of the 1.27x1.25 g/cc parent

	Parent	710x500	500x355	355x250	250x180	180x106	106x38
A	0.00	0.12	1.35	0.27	6.09	0.00	0.00
B	0.00	0.00	0.00	0.04	0.04	0.22	0.57
C	0.00	0.00	0.00	0.00	0.00	0.03	0.14
AB	54.96	34.93	40.28	46.35	31.09	33.83	49.25
AC	0.54	0.47	0.29	0.35	0.47	0.60	0.50
BC	0.00	0.00	0.00	0.00	0.00	0.02	0.03
ABC	44.50	64.48	58.08	53.00	62.30	65.30	49.51

Table 9 Measured liberation pattern of the 1.29x1.27 g/cc parent.

	Parent	710x500	500x355	355x250	250x180	180x106	106x38
A	0.00	0.00	0.00	0.02		0.29	0.43
B	0.00	0.00	0.00	0.00		0.00	0.36
C	0.00	0.00	0.00	0.00		0.00	0.18
AB	43.80	48.88	44.50	50.66		29.73	40.73
AC	0.61	0.56	0.49	0.45		0.68	0.38
BC	0.00	0.00	0.00	0.00		0.00	0.06
ABC	55.59	50.56	55.01	48.87		69.31	57.86

Table 10 Measured liberation pattern of the 1.31x1.29 g/cc parent.

	Parent	710x500	500x355	355x250	250x180	180x106	106x38
A	0.00	0.00	0.00	0.00	0.00	0.00	0.02
B	0.00	0.00	0.00	0.04	0.20	0.52	0.26
C	0.00	0.00	0.00	0.00	0.00	0.05	0.15
AB	23.97	37.84	43.70	47.08	19.50	27.36	38.21
AC	0.58	0.67	0.61	0.52	0.45	0.45	0.41
BC	0.00	0.00	0.00	0.00	0.00	0.01	0.04
ABC	75.44	61.49	55.69	52.36	79.85	71.61	60.90

Table 11 Measured liberation pattern of the 1.33x1.31 g/cc parent

	Parent	710x500	500x355	355x250	250x180	180x106	106x38
A	0.00	0.00	0.00	0.00	0.00	0.00	0.00
B	0.00	0.04	0.05	0.24	0.10	0.28	0.29
C	0.00	0.00	0.00	0.00	0.01	0.09	0.20
AB	15.94	25.22	10.80	30.62	37.40	22.99	46.52
AC	0.53	0.66	0.55	0.62	0.72	0.47	0.53
BC	0.00	0.03	0.00	0.00	0.01	0.04	0.10
ABC	83.53	74.04	88.60	68.51	61.76	76.12	52.36

Table 12 Measured liberation pattern of the 1.37x1.33 g/cc parent

	Parent	710x500	500x355	355x250	250x180	180x106	106x38
A	0.00	0.00	0.00	0.00	0.00	0.00	0.00
B	0.03	0.26	0.16	0.12	0.00	0.53	2.09
C	0.00	0.00	0.00	0.00	0.00	0.08	0.73
AB	24.66	6.47	26.91	31.20	40.36	17.58	10.96
AC	0.72	0.52	0.75	0.65	0.61	0.46	0.44
BC	0.00	0.00	0.00	0.00	0.00	0.03	0.33
ABC	74.60	92.75	72.19	68.03	59.03	81.32	85.45

Table 13 Measured liberation pattern of the 1.60x1.37 g/cc parent

	Parent	710x500	500x355	355x250	250x180	180x106	106x38
A	0.00	0.00	0.00	0.00	0.00	0.00	0.00
B	0.00	1.05	0.05	0.07	0.23	0.26	1.23
C	0.04	0.02	0.00	0.10	0.13	0.23	0.37
AB	1.16	36.96	24.12	9.41	28.43	32.10	42.28
AC	0.52	0.35	0.53	0.38	0.55	0.60	0.62
BC	0.00	0.06	0.00	0.00	0.11	0.13	0.52
ABC	98.28	61.56	75.30	90.03	70.54	66.68	54.98

Table 14 Measured liberation pattern of the +1.60 g/cc parent

	Parent	710x500	500x355	355x250	250x180	180x106	106x38
A	0.00	0.00	0.00	0.00	0.00	0.00	0.00
B	3.88	9.48	2.55	25.74	0.00	34.72	34.47
C	1.37	0.00	0.36	2.17	1.32	1.49	2.99
AB	0.77	50.62	8.17	4.49	0.28	19.87	11.94
AC	1.58	0.00	0.33	0.41	0.43	0.34	0.64
BC	29.37	0.00	4.74	15.92	24.84	4.53	12.19
ABC	63.03	39.90	83.84	51.27	73.14	39.06	37.77

The pattern of liberation of the three phases is unusual and interesting. The pattern generated by the +1.60 g/cc fraction is quite conventional and similar the pattern that is commonly observed with more conventional mineral systems. The parent particles are concentrated in the ternary ABC and binary BC classes. At this density there are of course comparatively few particles that contain coal. As size reduction proceeds there is a distinct reduction in the proportion of ternary ABC particles. (63% in the parent reducing to 38% at 106x38 μm). At the same time the binaries AB and AC increase significantly. The binary BC (ash-pyrite) decreases at small degrees of size reduction and then increases. The pure phases B and C increase significantly. No completely liberated coal is produced from the high density fraction even at high degrees of size reduction. The general pattern of liberation shows a consistent transfer from the ternaries to binaries and liberated single-phase particles.

Figure 32 Variation of progeny density with size.

At the other end of the scale the pattern is essentially different. Particles that contain very little ash or pyrite show a distinct tendency to fracture in such a way that the proportion of binaries and ternaries *increases* as the progeny size decreases. This behavior, of course, runs counter to simple intuitive models of liberation processes which assert that liberation inevitably increases as size decreases. However it is widely observed that ash-forming minerals tend to concentrate in the finer sizes of fine coal samples. This is due to non-random fracture that occurs when coal particle are comminuted. Although the coal phase is undoubtedly softer than most ash-forming minerals and pyrite, the latter are more brittle and consequently fracture more easily under the kind of impact stress the occurs in ball mills and other high intensity impact mills such as the small hammer mill that was used in this study to perform the comminution. Concentration of unliberated particles at the

smaller sizes during comminution is symptomatic of enhanced fracture in the region of grain boundaries and was first observed by King and Middlemiss (1998). This fragmentation behavior of coal has potentially important implications for the design of comminution and de-ashing - desulfurizing circuits for the production of pulverized coal fuel. Maximum removal of these troublesome components can be achieved by a sensible combination of comminution and separation operations. Optimum removal is unlikely to be achieved by conventional sequential comminution followed by separation. Grain boundary region fracture has the really desirable feature of concentrating unliberated material in the finer sizes even at only modest overall degrees of comminution and this effect should be exploitable to enhance overall comminution and coal cleaning efficiency.

The data presented in Tables 7 to 14 provides the necessary quantitative information that can be used in a comprehensive simulator such as MODSIM. This popular simulator was equipped, during the course of this project, with a number of models for comminution operations that accurately account for liberation of multi-component materials. This simulator has been shown to be effective for the optimization of coal washing plants. (King, 1999)

6 A LIBERATION MODEL FOR COAL

The successful development of the imaging procedure for coal made it possible to generate the raw data from which a model of the liberation characteristics of the three phases in coal can be developed that is independent of the specific comminution process that is used to generate the fine particles. This model is based on the linear stochastic method that is described in King (1994). Linear intercept analysis has turned out to be a very effective tool for the study of mineral liberation. Linear intercepts can be measured easily, accurately and unambiguously using standard image-analysis equipment so that good and reliable data generated from measurements on real ore minerals are available to calibrate and test theoretical models that are used to calculate, predict and measure liberation in situations that are met in industrial operations. Linear intercept data can be considered to be a one-dimensional sampling of three-dimensional mineralogical textures and particles. This sampling is generated by a line probe that penetrates the material. Line-probe data can be easily obtained in practice by sectioning followed by linear scanning along a random line on the section.

Ease and accuracy of measurement is not the only advantage in using linear intercept data. Since the linear-probe line will pass successively through the different mineral phases, the development of the signal from the probe can be converted to a stepwise function that steps from one phase to another at points on the scan line that are associated with a phase boundary. Since the phase boundaries that are intersected are positioned randomly along the scan line, the probe output signal can be modeled

by a 3-state one-dimensional stochastic process that develops in distance along the scan line. The advantage in adopting this point of view is that a large body of results is available for one-dimensional stochastic processes, which have been extensively studied. This extensive collection provides results and theorems that can be used directly to generate workable and useful solutions to many of the problems in mineral liberation analysis. This approach to the study of mineral liberation was first exploited by King in 1979 who showed that the entire linear grade distribution after random breakage of a two-phase mineral texture can be predicted from a knowledge of the linear intercept distributions through the two phases in the unbroken ore. In spite of the apparent difficulty associated with this problem, the result could be obtained easily by making some simple extensions to a theorem originally proved by Takács (1957) for alternating renewal processes.

An important extension to multiphase systems (more than two phases) recently developed by

Figure 33 Image of unbroken coal sample used to measure distribution of linear intercept lengths for each of the phases. Coal phase is black, ash minerals are grey and pyrite is white.

Yingling (1991) is used as the basis for the analysis of the three phase system that is studied here. The main mathematical results that are used here have been developed by Gibson and Conolly (1971).

Figure 34 Image of unbroken coal on which the distribution of linear intercept lengths for each of the phases were measured. Coal is black, ash minerals are grey and pyrite is white.

This model is based on the idea of sampling the whole ore texture by means of a linear probe that penetrates the ore and registers the alternating pattern of intercepts through the three phases that make up the ore. The fundamental data is the distribution of intercept lengths for each of the phases which are generated from a large number (many thousands) of intercepts through each phase. In practice this probe is generated from a succession of linear probe lines drawn on images of samples of the unbroken ore obtained in the SEM. Two typical such images from samples of Pittsburgh#8-1997 are shown in Figures 33 and 34. In all, 80 such images were obtained using polished samples approximately 1 inch square which were cut at different orientations from a single "typical" large lump of the coal sample. Although this did not produce a representative sampling of the entire coal, it did produce typical distributions of linear intercepts for each of the three phases and typical

transition probabilities among the three phase. It should be pointed out that the images reproduced here do not show the full detail that was obtained because of the relatively poor resolution of the laser printer used for reproduction.

6.1 Distribution of Linear Incept Lengths for the Phases

The measured distribution of linear intercept lengths for each of the phases are shown in Figure 35 and the matrix of transition probabilities are given in Table 15.

Table 15 Transition probabilities of all phases in unbroken coal Pittsburgh #8 1997

	Ash	Pure coal	Pyrite
Ash	0	0.4715	0.0153
Pure coal	0.4716	0	0.0131
Pyrite	0.0150	0.0134	0

A cumulative exponential sum distribution function that was found to fit the measured data well for each of the phases.

$$F(l) = 1 - \sum_{i=1}^m \alpha_i e^{-\lambda_i l} \quad (7)$$

where m is a positive integer and α_i and λ_i are real parameters with $\sum_{i=1}^m \alpha_i = 1$.

The density distribution corresponding to $F(t)$ will be

$$f(l) = \sum_{i=1}^m \alpha_i \lambda_i e^{-\lambda_i l} \quad (8)$$

Least squares values for the parameters were obtained for each of the three phases using the measured data. The results for pure coal, pyrite and other ash-forming minerals can be described by the following equations, respectively:

$$F_c(l) = 1 - 0.4928e^{-0.1274l} - 0.1605e^{-19.8091l} - 0.3467e^{-0.0156l} \quad (9)$$

$$F_p(l) = 1 - 0.8126e^{-0.3392l} - 0.1874e^{-0.0602l} \quad (10)$$

$$F_a(l) = 1 - 0.9326e^{-0.5511l} - 0.0674e^{-0.0602l} \quad (11)$$

Figure 35 Measured cumulative linear length distributions for pure coal, pyrite and other ash-forming minerals in unbroken coal Pittsburgh #8 1997

The graphs corresponding to Equations 9 to 11 and the discrete linear intercept length distribution data are plotted in Figure 35. Those graphs show that the fitted equations describe their corresponding data well.

In the process of this research, linear intercept length distributions for many other ore systems were also tested. All fitted results indicated that linear intercept length distribution data could be well expressed by just several exponentials, at most four exponentials, which was actually unusual.

6.2 Calculation of the Linear Grade Distribution after Comminution

The sample linear probe is considered to be made up of a succession of phase intercepts laid head to tail, in random order and each having random length taken from the distributions (9) - (11). The sequencing must satisfy the transition probabilities given in Table 15. The strategy used in the linear stochastic model for the prediction of liberation is to calculate the probability of occurrence

of every linear intercept type when the probe line is broken to produce a distribution of fragment lengths that is equivalent to the linear transects across the particles that are produced by comminution.

The direct calculation of the probabilities of occurrence of all possible linear intercept types is cumbersome to say the least and some assumptions must be made to limit the number of linear types that must be considered in the theoretical model. The porphyry model that was developed for three-phase textures in King (1994) is not a suitable for coal. This model assumes that two of the phases in the texture are sufficiently dispersed to preclude the observation of linear intercepts from the particle sections that contain more the two segments of these phases. A glance at Figures 33 and 34 confirms that this assumption is far too restrictive to provide a reasonable description of the coal particles. An alternative model was accordingly investigated. This model was based on an analysis of three-state sojourn time problems that was presented by Gibson and Conolly (19971).

For fragments that contain all three phases

$$\begin{aligned}
 &P_{AB_1B_2}(g_A, g_{B_1}|l) = \\
 &\nu_A \{p(g_A, g_{B_1}, TE=A|LE=A, l) + p(g_A, g_{B_1}, TE=B_1|LE=A, l) + p(g_A, g_{B_1}, TE=B_2|LE=A, l) + \\
 &\nu_{B_1} \{p(g_A, g_{B_1}, TE=A|LE=B_1, l) + p(g_A, g_{B_1}, TE=B_1|LE=B_1, l) + p(g_A, g_{B_1}, TE=B_2|LE=B_1, l) + \\
 &\nu_{B_2} \{p(g_A, g_{B_1}, TE=A|LE=B_2, l) + p(g_A, g_{B_1}, TE=B_1|LE=B_2, l) + p(g_A, g_{B_1}, TE=B_2|LE=B_2, l)
 \end{aligned} \tag{12}$$

where TE represents the trailing edge of a fragment length and LE represents the leading edge of a linear fragment. g_A and g_{B_1} represent the grades of coal and ash respectively and l is the length of the intercept through the particle. These separate probability densities are evaluate using the results of Gibson and Conolly to give

$$\begin{aligned}
 &p(g_A, g_{B_1}, TE=A|LE=A, l) = \\
 &\frac{1}{\mu_A} \sum_{n=1}^{\infty} \sum_{m=n+1}^{\infty} \alpha_1^n \alpha_2^{m-n} \binom{m}{n} h_1^{(n)}(g_{B_1} l) h_2^{(m-n)}(l - g_A l - g_{B_1} l) \int_0^{g_A l} \{G^{(m-1)}(t) - 2G^{(m)}(t) + G^{(m+1)}(t)\} dt \tag{13}
 \end{aligned}$$

$$\begin{aligned}
 &p(g_A, g_{B_1}, TE=B_1|LE=A, l) = \\
 &\frac{1}{\mu_A} \sum_{n=1}^{\infty} \sum_{m=n+1}^{\infty} \alpha_1^n \alpha_2^{m-n} \binom{m-1}{n-1} \{G^{(m-1)}(g_A l) - G^{(m)}(g_A l)\} h_2^{(m-n)}(l - g_A l - g_{B_1} l) \{H_1^{(n-1)}(g_{B_1} l) - H_1^{(n)}(g_{B_1} l)\} \tag{14}
 \end{aligned}$$

$$p(g_A, g_{B_1}, TE=B_2|LE=A, l) =$$

$$\frac{1}{\mu_A} \sum_{n=1}^{\infty} \sum_{m=n+1}^{\infty} \alpha_1^n \alpha_2^{m-n} \binom{m-1}{n} \{G^{(m-1)}(g_A l) - G^{(m)}(g_A l)\} h_1^{(n)}(g_{B_1} l) \\ \{H_2^{(m-n-1)}(l - g_A l - g_{B_1} l) - H_2^{(m-n)}(l - g_A l - g_{B_1} l)\} \quad (15)$$

$$p(g_A, g_{B_1}, TE=A | LE=B_1, l) =$$

$$\frac{1}{\mu_{B_1}} \sum_{n=0}^{\infty} \sum_{m=n+1}^{\infty} \alpha_1^n \alpha_2^{m-n} \binom{m}{n} \{H_1^{(n)}(g_{B_1} l) - H_1^{(n+1)}(g_{B_1} l)\} h_2^{(m-n)}(l - g_A l - g_{B_1} l) \{G^{(m)}(g_A l) \\ - G^{(m+1)}(g_A l)\} \quad (16)$$

$$p(g_A, g_{B_1}, TE=B_1 | LE=B_1, l) =$$

$$\frac{1}{\mu_{B_1}} \sum_{n=1}^{\infty} \sum_{m=n+1}^{\infty} \alpha_1^n \alpha_2^{m-n} \binom{m-1}{n-1} g^{(m)}(g_A l) h_2^{(m-n)}(l - g_A l - g_{B_1} l) \int_0^{g_{B_1} l} \{H_1^{(n-1)}(t) - 2H_1^{(n)}(t) + H_1^{(n+1)}(t)\} dt \quad (17)$$

$$p(g_A, g_{B_1}, TE=B_2 | LE=B_1, l) =$$

$$\frac{1}{\mu_{B_1}} \sum_{n=0}^{\infty} \sum_{m=n+1}^{\infty} \alpha_1^n \alpha_2^{m-n} \binom{m-1}{n} g^{(m)}(g_A l) \{H_1^{(n)}(g_{B_1} l) - H_1^{(n+1)}(g_{B_1} l)\} \\ \{H_2^{(m-n-1)}(l - g_A l - g_{B_1} l) - H_2^{(m-n)}(l - g_A l - g_{B_1} l)\} \quad (18)$$

$$p(g_A, g_{B_1}, TE=A | LE=B_2, l) =$$

$$\frac{1}{\mu_{B_2}} \sum_{n=1}^{\infty} \sum_{m=n}^{\infty} \alpha_1^n \alpha_2^{m-n} \binom{m}{n} h_1^{(n)}(g_{B_1} l) \{H_2^{(m-n)}(l - g_A l - g_{B_1} l) - H_2^{(m-n+1)}(l - g_A l - g_{B_1} l)\} \\ \{G^{(m)}(g_A l) - G^{(m+1)}(g_A l)\} \quad (19)$$

$$p(g_A, g_{B_1}, TE=B_1 | LE=B_2, l) =$$

$$\frac{1}{\mu_{B_2}} \sum_{n=1}^{\infty} \sum_{m=n}^{\infty} \alpha_1^n \alpha_2^{m-n} \binom{m-1}{n-1} g^{(m)}(g_A l) \{H_2^{(m-n)}(l-g_A l-g_{B_1} l) - H_2^{(m-n+1)}(l-g_A l-g_{B_1} l)\} \\ \{H_1^{(n-1)}(g_{B_1} l) - H_1^{(n)}(g_{B_1} l)\} \quad (20)$$

$$p(g_A, g_{B_1}, TE=B_2 | LE=B_2, l) = \\ \frac{1}{\mu_{B_2}} \sum_{n=1}^{\infty} \sum_{m=n+1}^{\infty} \alpha_1^n \alpha_2^{m-n} \binom{m-1}{n} g^{(m)}(g_A l) h_1^{(n)}(g_{B_1} l) \int_0^{l-g_A l-g_{B_1} l} \{H_2^{(m-n-1)}(t) - 2H_2^{(m-n)}(t) \\ + H_2^{(m-n+1)}(t)\} dt \quad (21)$$

For intercepts that include only two phases

$$p_{AB_i}(g_A | l) = v_A \{p(g_A, TE=A | LE=A) + p(g_A, TE=B_i | LE=A)\} \\ v_{B_i} \{p(g_A, TE=A | LE=B_i) + p(g_A, TE=B_i | LE=B_i)\} \quad (22)$$

where i can be 1 or 2.

$$p(g_A, TE=A | LE=A, l) = \frac{1}{\mu_{A,m=1}} \sum_{m=1}^{\infty} \alpha_i^m h_i^{(m)}(l-g_A l) \int_0^{g_A l} \{G^{(m-1)}(t) - 2G^{(m)}(t) + G^{(m+1)}(t)\} dt \quad (23)$$

$$p(g_A, TE=B_i | LE=A, l) = \frac{1}{\mu_{A,m=1}} \sum_{m=1}^{\infty} \alpha_i^m \{G^{(m-1)}(g_A l) - G^{(m)}(g_A l)\} \{H_i^{(m-1)}(l-g_A l) - H_i^{(m)}(l-g_A l)\} \quad (24)$$

$$p(g_A, TE=A | LE=B_i, l) = \frac{1}{\mu_{B_i,m=0}} \sum_{m=0}^{\infty} \alpha_i^m \{H_i^{(m)}(l-g_A l) - H_i^{(m+1)}(l-g_A l)\} \{G^{(m)}(g_A l) - G^{(m+1)}(g_A l)\}$$

(25)

$$p(g_A, TE=B_i | LE=B_i, l) = \frac{1}{\mu_{B_i, m=1}} \sum_{m=1}^{\infty} \alpha_i^m g^{(m)}(g_A l) \int_0^{l-g_A l} \{H_i^{(m-1)}(t) - 2H_i^{(m)}(t) + H_i^{(m+1)}(t)\} dt$$

(26)

For fragments that consist of a single phase:

$$Pr\{pure A | l\} = v_A \left\{ 1 - \int_0^l \frac{1-G(t)}{\mu_A} dt \right\}$$

(27)

$$Pr\{pure B_i | l\} = v_{B_i} \left\{ 1 - \int_0^l \frac{1-H_i(t)}{\mu_{B_i}} dt \right\}$$

(28)

where i can be 1 or 2.

In these equations, superscripts such as (m) represent the m -fold convolution of the associated function. G represents the cumulative distribution of linear intercepts through the coal phase in the unbroken ore and g its associated density. H_1 represents the cumulative distribution of linear intercepts through the ash phase in the unbroken ore and h_1 its associated density. H_2 represents the cumulative distribution of linear intercepts through the pyrite phase in the unbroken ore and h_2 its associated density.

Equations 12 to 28 form the basis of the predictive model. They allow the calculation of the probabilities of occurrence of each of the 43 linear grade classes that define the ternary system. They are given entirely in terms of the linear intercept distributions of the three separate phases in the

unbroken ore and in terms of the interphase transition probabilities. The prediction is completed by applying the stereological correction to the calculated linear distribution of grades. Although these equations appear to be unduly complex, they are surprisingly easy to use. The main difficulty is the evaluation of the m-fold convolutions of the distribution functions. Since these distributions were found to be sums of exponentials, the general expressions for evaluating their m-fold convolutions were developed. These are described in appendix 1.

7 CONCLUSIONS

1. A method has been developed to produce detailed images of coal samples by SEM back-scattered electron detection. Images of coal particles are suitable for the measurement of the liberation spectrum with respect to the three phases: coal, ash-forming minerals, and pyrite. Images of 1" x 1" sections are suitable for the determination of the linear intercept distributions for these three phases in the unbroken coal.
2. The three phases of primary interest can be accurately delineated in the images.
3. Separate minerals can be identified using a new calibration method based on the mass-weighted average atomic number of the mineral.
4. The quantities of each phase in the images can be accurately measured using the area fraction. The density of any sample of coal can be estimated to a precision approximately 10 kg/m³ using the measured area fractions and the known densities of the phases.
5. The disposition of ash in the progeny particles that result from the comminution of 1 mm coal particles was determined using the dense-liquid method, and more comprehensively using the image analysis method, which was shown to be sufficiently efficient and accurate to determine the liberation spectrum for each phase in all the progeny particles.

6. A restricted set of seven particle types was considered. These include the three separate phases, all binaries collected into three classes and all ternaries in a single class. The matrix of cross-transfer coefficients for these particle types was determined in a form that can be used as an Andrews-Mika diagram. Although comparatively crude, this formulation can be used to simulate the disposition of ash and pyrite when coal is subjected to comminution.
7. A model for the liberation of coal using a stochastic linear representation of the unbroken ore was developed based on a three-state renewal process. This model was found to be computationally tractable and predicted the linear grade distribution for 43 particle types in the three-phase space. A straightforward stereological procedure was used to convert these predictions into the distribution by volume at any particle size.

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Appendix 1 Calculation of N-Fold Convolutions of Exponential Distributions

In order to make effective use of the liberation model it is necessary to calculate the n-fold convolutions of the distribution density functions that were measured for the individual phases in the unbroken coal. These distributions were found to be well described by sums of exponentials and a general algorithm was developed for this purpose.

In many problems of probability theory and stochastic process, one quite often has to calculate the n-fold convolution of distribution functions with themselves. In general, the calculating formulae of n-fold convolution are only available for some very simple function forms, such as one-exponential functions^[1]. Lasser and Tritschler developed a set of recursive formulae of n-fold convolution for a type of special elementary functions^[2]. Here, we are going to exploit the general analytical solution for the n-fold convolution of distribution functions in exponential-sum forms with themselves.

In practice, distribution functions are usually measured by experiments and then the measured data are fitted to some function forms. Fortunately, it has been long known that experimental data can fit to some exponential sums under controlled errors.. Using exponential-sum fitting, a density distribution function can be expressed by

$$f(t) = \sum_{i=1}^m a_i e^{-b_i t}$$

Where, m , a finite integer, is the number of exponentials, a_i and b_i are non-negative constants and we shall just consider that $f(t)$ is defined in (1) when $t \in [0, \infty)$ and $f(t) = 0$ when $t < 0$.

By definition, the n-fold convolution of $f(t)$ with itself is $f^{(n)}(t) = f * f * \dots * f(t)$ while the n-fold convolution of cumulative distribution function corresponding to $f^{(n)}(t)$ is

$$F^{(n)}(t) = \int_0^t f^{(n)}(x) dx . \text{ Employing Laplace transform, it is easily acquired}$$

$$(2) f^{(n)}(t) = L^{-1} [(L[f(t)])^n]$$

and

$$(3) F^{(n)}(t) = L^{-1} \left[\frac{L[f^{(n)}(t)]}{s} \right]$$

Where, L and L^{-1} represent forward and inverse Laplace transforms, respectively.

Therefore, our mission here is to find the solution for (2) and (3) provided that $f(t)$ is defined by (1).

2. Derivation of $f^{(n)}(t)$ and $F^{(n)}(t)$

First of all, let us prove a preliminary theorem.

Theorem 1 If function $H(x)$ is defined by $H(x) = \frac{1}{\prod_{i=1}^m (x+c_i)^{n_i}}$, where m , n_i are

nonzero positive integers, c_i is a constant and $x \in (-\infty, +\infty)$, but $x \neq -c_i$, its n th order derivative can be calculated by

$$(4) \frac{d^n H(x)}{dx^n} = \sum_{i=1}^n (-1)^{n-i+1} \frac{(n-1)!}{(i-1)!} G_{(n-i+1)}(x) \frac{d^{n-1} H(x)}{dx^{n-1}}$$

where $G_{(k)}(x)$ is defined by

$$(5) \quad G_{(k)}(x) = \sum_{i=1}^m \frac{n_i}{(x+c_i)^k}$$

Proof (4) is obviously correct for $n = 1$. Let us assume (4) is correct for $n = N$.

Therefore

$$\frac{d^N H(x)}{dx^N} = \sum_{i=1}^N (-1)^{N-i+1} \frac{(N-1)!}{(i-1)!} G_{(N-i+1)}(x) \frac{d^{N-1} H(x)}{dx^{N-1}}$$

Do once derivative to above equation and notice $\frac{dG_{(k)}(x)}{dx} = -kG_{(k+1)}(x)$

$$\frac{d^{N+1} H(x)}{dx^{N+1}} = \sum_{i=1}^N [(-1)^{N-i} \frac{(N-1)!}{(i-1)!} (N-i+1) G_{(N-i+2)}(x) \frac{d^{N-1} H(x)}{dx^{N-1}} + (-1)^{N-i+1} \frac{(N-1)!}{(i-1)!} G_{(N-i+1)}(x) \frac{d^i H(x)}{dx^i}]$$

Expanding above equation and combining all similar items, we immediately have

$$\frac{d^{N+1} H(x)}{dx^{N+1}} = \sum_{i=1}^{N+1} (-1)^{N+1-i+1} \frac{(N+1-1)!}{(i-1)!} G_{(N+1-i+1)}(x) \frac{d^{N+1-i} H(x)}{dx^{N+1-i}}$$

Therefore, by induction we can say for any $n > 0$, (4) is all correct. As the result, theorem 1 is proven.

Let us define a short symbol to represent some consecutive summations.

Definition

$$1 \sum_{\langle m, n, p \rangle} = \sum_{i_1=1}^{m-(p-1)} \sum_{i_2=i_1+1}^{m-(p-2)} \sum_{i_3=i_2+1}^{m-(p-3)} \dots \sum_{i_p=i_{p-1}+1}^m \sum_{n_1=1}^{n-(p-1)} \sum_{n_2=1}^{n-n_1-(p-2)} \dots \sum_{n_p=1}^{n-\sum_{i=0}^{p-2} n_i-1} \sum_{n_p=n-\sum_{i=0}^{p-1} n_i}^{n-\sum_{i=0}^{p-1} n_i}$$

From now on we shall present the solution for (2) and (3) in the following theorems.

Theorem 2 Provided that $f(t)$ is defined by (1), the n-fold convolution of $f(t)$ can be calculated by

$$(5) f^{(n)}(t) = \sum_{p=1}^m \sum_{\langle m, n, p \rangle} \binom{n}{n_1 n_2 \dots n_p} \prod_{q=1}^p a_{i_q}^{n_q} \sum_{k=1}^p \sum_{T=1}^{n_k} \frac{\Phi_{kT}^{(p)}(-b_{i_k})}{(n_k-T)!(T-1)!} t^{n_k-T} e^{-b_{i_k}t}$$

where

$$(6) \Phi_{kT}^{(p)}(s) = \frac{d^{T-1}}{ds^{T-1}} \left[\frac{(s+b_{i_k})^{n_k}}{\prod_{q=1}^p (s+b_{i_q})^{n_q}} \right]$$

Proof Perform Laplace transform to (1) and take the corresponding result into (2)

$$f^{(n)}(t) = L^{-1} \left[\left(\sum_{i=1}^m \frac{a_i}{s+b_i} \right)^n \right]$$

Apply multinomial expansion theorem^[9] to above equation

$$f^{(n)}(t) = L^{-1} \left[\sum \binom{n}{n_1, n_2, \dots, n_m} \left(\frac{a_1}{s+b_1} \right)^{n_1} \left(\frac{a_2}{s+b_2} \right)^{n_2} \dots \left(\frac{a_m}{s+b_m} \right)^{n_m} \right]$$

where, the sum is over all m - tuples of nonnegative integers, $n_1, n_2, \dots, n_m$ that sum to

n .

Making use of definition 1, above equation can be expressed by

$$f^{(n)}(t) = \sum_{p=1}^m \sum_{\langle m, n, p \rangle} \binom{n}{n_1, n_2, \dots, n_p} \prod_{q=1}^p a_{i_q}^{n_q} L^{-1} \left[\frac{1}{\prod_{q=1}^p (s+b_{i_q})^{n_q}} \right]$$

From the table of Laplace transforms^[10], $L^{-1} \left[\frac{1}{\prod_{q=1}^p (s+b_{i_q})^{n_q}} \right]$ can be found. Take the result into

above equation and we can reach (5) immediately. Therefore, the proof is finished.

Theorem 3 Provided that $f(t)$ is defined by (1), the n -fold convolution of $F(t)$ can be calculated by

$$(7) \quad F^{(n)}(t) = 1 - \sum_{p=1}^m \sum_{\langle m, n, p \rangle} \binom{n}{n_1, n_2, \dots, n_p} \prod_{q=1}^p a_{i_q}^{n_q} \sum_{k=1}^p \sum_{T=1}^{n_k} \frac{\Phi_{kT}^{(p)}(-b_{i_k})}{(T-1)!} b_{i_k}^{-(n_k-T+1)} e^{-b_{i_k} t} \sum_{r=0}^{n_k-T} \frac{(b_{i_k} t)^r}{r!}$$

where $\Phi_{kT}^{(p)}(s)$ is defined by (6).

Proof Taking (5) into (3) and alternatively doing forward and inverse Laplace transform operations, $F^{(n)}(t)$ is expressed by

$$F^{(n)}(t) = \sum_{p=1}^m \sum_{\langle m, n, p \rangle} \binom{n}{n_1, n_2, \dots, n_p} \prod_{q=1}^p a_{i_q}^{n_q} \sum_{k=1}^p \sum_{T=1}^{n_k} \frac{\Phi_{kT}^{(p)}(-b_{i_k})}{(T-1)!} b_{i_k}^{-(n_k-T+1)} [1 - e^{-b_{i_k}t} \sum_{r=0}^{n_k-T} \frac{(b_{i_k}t)^r}{r!}]$$

Because $\lim_{t \rightarrow \infty} F^{(n)}(t) = 1$, above equation can be easily simplified to (7). Consequently,

theorem3 is proven.

The remaining thing is to calculate $\Phi_{kT}^{(p)}(s)$.

Theorem 4 If $p = 1$, $\Phi_{1T}^{(p)}(s) = 1$ at $T = 1$ and $\Phi_{1T}^{(p)}(s) = 0$ elsewhere; otherwise if

$p \geq 2$, there exists the following calculating formula

$$(8) \quad \Phi_{kT}^{(p)}(s) = \sum_{i=1}^{T-1} (-1)^{T-i} \frac{(T-2)!}{(i-1)!} G_{(T-i)}(s) \Phi_{k, i-1}^{(p)}(s)$$

where

$$(9) \quad G_{(T-i)}(s) = \sum_{q=1, q \neq k}^p \frac{n_q}{(s+b_{i_q})^{T-i}}$$

$$(10) \quad \Phi_{kl}^{(p)}(s) = \frac{1}{\prod_{q=1, q \neq k}^p (s+b_{i_q})^{n_q}}$$

and k is defined in (7)

Proof Notice $1 \leq k \leq p$. Obviously $\Phi_{11}^{(1)}(s)=1$ and $\Phi_{1T}^{(1)}(s)=0$ for $T>1$.

Applying theorem 1 to (6), we can immediately reach (8), (9) and (10) for $p \geq 2$. Therefore, theorem 4 is proven.

3 An Example

In mineral processing engineering, the linear intercept length distribution for each component in unbroken ores can be measured by automatic image analysis coupled with scanning electronic microscopy. The measured data can be easily fitted to exponential sums. The n-fold convolution of linear intercept length distribution has to be calculated in order to predict the liberation spectra of minerals from ores^[11].

Here we have a two component Brazil ore, the linear intercept length distributions of whose mineral and gangue are described by the following exponential sum models, respectively

$$(11) \quad f_m(t) = 0.2037e^{-0.2170t} + 0.00378e^{-0.0616t}$$

$$(12) \quad f_g(t) = 0.190420e^{-0.2604t} + 0.002933e^{-0.0165t} + 0.000073e^{-0.0008t}$$

Figure 36 N-fold convolution of linear intercept length distribution for mineral phase in a Brazil ore

Using theorems 2, 3 and 4, the n -fold convolution of $f(t)$ and $F(t)$ is calculated and plotted in Figures 1 and 2.

Figure 37 N-fold convolution of linear intercept length distribution for gangue phase in a Brazil ore