STATE OF ILLINOIS
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VARIATION IN COAL REFLECTANCE

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CIRCULAR 241 1957

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ABSTRACT

This investigation was undertaken in order to study in detail the minor variations of coal reflectance, such as that within and between vitrain bands in a block of coal, between blocks of coal, and between vitrinites, fusinites, and semi-fusinites in the same coal.

Reflectance was measured with an electronic photometer and photomultiplier search unit. Calibration of standards, quartz and synthetic ruby, established the accuracy of measurement. Using statistical tests (analysis of variance) as a guide, experimental procedures were chosen to assure a minimum of experimental error.

Variation of vitrinites with rank seems to be essentially continuous and the results are consistent with those of Broadbent and Shaw (1955). Variation within vitrain bands is negligible. Variation between some vitrain bands in a block of coal is significant. Variation between vitrain bands of blocks of the same coal from one locality is less than that between vitrain bands within the blocks. A study of the variation in reflectance of vitrinites, semi-fusinites, and fusinites, and micrinites in individual coal samples shows that there is a continuum in values for all of these components.

INTRODUCTION

During the past decade many of the attempts to elucidate the chemical and physical structure of coal have been centered on the measurement and interpretation of physical properties of the coal substance, including, among others, optical properties, density, internal surface, hardness, and magnetic susceptibility. The study of optical properties has been concerned mainly with reflectance, which, among other methods (McCabe, 1937), can be used as an indirect measurement of the index of refraction.

Most of the investigations of coal reflectance have stressed the relationship between rank, or metamorphic grade, and the reflecting power of vitrains or vitrinites. The earliest work that pointed out the rise in reflectance with increasing rank of coal was that of Hoffman and Jenkner (1932). For more than ten years following, there was little activity in the field, but in 1943, Seyler (1943) reopened the subject and proposed a theory of reflectance based on a stepwise, discontinuous rise of reflecting power with increase in rank (as measured by carbon content).

Seyler included in ten steps the range of all ranks of coal, and implicit in his ideas was the conclusion that chemical and structural changes took place by discrete jumps during coal metamorphism. To Seyler must go the credit for bringing reflectance to the forefront as a valuable tool in unravelling the complexities of coal structure and as a rapid physical method for determining the grade of coal metamorphism.

During the years since the appearance of Seyler's theory many other workers have published investigations of coal reflectance (see list of references). Some of them have accepted, with modification, Seyler's theory as originally stated

(Mukherjee, 1952; Huntjens and van Krevelen, 1954). Others have rejected it (Dahme and Mackowsky, 1950; McCartney, 1952). There also has been a wide divergence of opinion on the appropriateness of the Berek microphotometer (used by Hoffman and Jenkner, Seyler, and others) as compared with a photocell and galvanometer or electronic photometer for studies of coal reflectance. Some workers have measured only vitrains of different ranks; others have studied many other coal components as well.

In the writer's opinion the controversy on the stepwise nature of increase in reflectance of vitrains has been stilled by the recent work of Broadbent and Shaw (1955). They have been the first to use modern methods of statistical analysis (in this case the lumped analysis of variance) in the interpretation of reflectance data. Their use of statistical methods in the analysis of a large amount of carefully accumulated data has demonstrated that reflectance varies more or less continuously with rank. The experimental data here presented that are pertinent to the problem corroborate their conclusions.

In previous investigations much effort has been spent on the major variation of reflectance with rank of coal and but little time has been spent on assessing more minute variations. It is the purpose of this paper to explore some of these variations in vitrinites, semi-fusinites, and fusinites, as well as to present some additional data on increase in reflectance with rank.

EXPERIMENTAL PROCEDURE

The equipment used in this investigation was a photomultiplier search unit and electronic photometer (Photovolt Corporation Model 512) attached to a binocular metallurgical microscope. The photometer search unit was placed over one (8X) ocular, and the paired ocular was left free for visual observation of the material being measured. A centered pinhole diaphragm, 0.025 inch in diameter, was placed at the position of the field diaphragm of the photometer ocular in order to greatly restrict the field of view so that reflectance readings of very small homogeneous areas could be made. The actual diameter of the field of view measured, of course, depends on the objective used. Other than the primary calibration of standards, all measurements were taken with oil immersion objectives (25X and 80X). It was because of the small field of view and the use of oil immersion objectives that a photomultiplier photometer was necessary.

In order to insure a minimum of experimental error, adjustments of the photometer zero and the vertical illuminator of the microscope were checked before each series of measurements. All measurements were made with the field diaphragm of the vertical illuminator stopped down as far as possible so as to have the narrowest possible cone of light impinging on the polished section and thus only the vertically incident light. The incandescent lamp used for illumination was connected to a constant voltage transformer (plus or minus 0.1 volt) and was allowed to warm up for at least one hour to minimize fluctuations in light levels.

The fundamental standard of reflectance used was a polished surface of quartz cut normal to the c-axis (the 0001 face). The secondary standard for use with oil immersion objectives was a polished synthetic ruby (corundum). Both quartz and ruby had lower faces cut at high angles to eliminate secondary reflections. The ruby was first calibrated in air with reference to the quartz and then used as the standard by which coal reflectance measurements were made. The measured reflectance of the ruby was 1.6 times that of quartz (in air) taking the reflectance value of quartz as 4.6 percent, the reflectance of ruby, as experimentally determined (in red light-650 millimicrons), as 7.4 percent. This value was checked by calculating the percentage of reflectance from the Fresnel equation:

$$R = \frac{(n-1)^2}{(n+1)^2} \times 100$$

where R is reflectance in percent and n is the index of refraction. The index of refraction of the ruby was determined by use of immersion liquids to be 1.75 (ω). The experimentally determined and calculated values of R agree, indicating that reflectance determinations using this equipment are accurate.

Polished surfaces of all samples of coal used in this study were prepared by uniform methods. The steps followed in the polishing routine were: (1) hand grinding on 3/0 emery paper, (2) lap grinding with 1μ diamond paste on a fine cotton cloth, (3) lap polishing with 1μ alumina on cotton cloth, and (4) hand polishing with wet billiard cloth. Only blocks of coal were used although some of the more friable specimens were mounted in lucite before polishing. All surfaces were measured with the same orientation relative to the microscope axis to eliminate anisotropy effects.

Samples used for reflectance determinations were splits from larger samples used for chemical analyses. Fixed carbon (unit-coal-basis - dry-mineral-matter free) and percentage of carbon (dry ash-free) for the samples used are shown in table 1. For complete proximate and ultimate analyses of these samples see Siever (1952, p. 345).

Table 1. - Partial Chemical Analyses of Samples

Sample	Fixed Carbon (unit coal basis) %	Carbon (dry-ash-free) %
Anthracites		
Mammoth, Pennsylvania	95.5	93.28
Holmes, Pennsylvania	94.8	91.82
Buck Mountain, Pennsylvania	93.3	91.45
Semi-anthracites		
Merrimac, Virginia	92.0	91.49
Low-volatile bituminous		
Pocahontas No. 6, West Virginia	82.0	90.44
Bakerstown, West Virginia	80.7	90.37
Brookville, Pennsylvania	80.7	89.54
Lower Kittanning, Pennsylvania	80.2	88.02
Pittsburgh, West Virginia	79.9	87.22
Medium-volatile bituminous		
Lower Kittanning, Pennsylvania	76.0	89.81
Lower Freeport, Pennsylvania	73.4	88.45
High-volatile bituminous A		
High Splint, Kentucky	61.3	84.44
Jellico, Tennessee	61.0	84.13
Alma, Kentucky	59.4	84.38
Amburgy, Kentucky	59.0	83.30
River Gem, Tennessee	58.9	83.91
Hazard No. 4, Kentucky	58.7	83.19
Hazard No. 6, Kentucky	58.5	83.43
Pewee, Tennessee	56.1	84.52

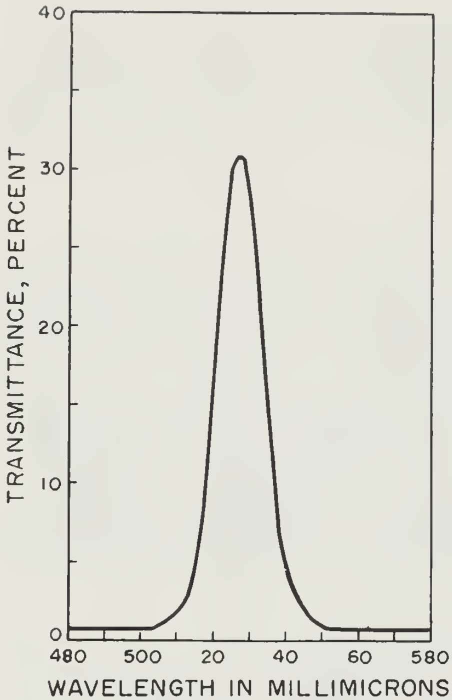

Fig. 1. - Spectral transmission of G.A.B. green filter.

All measurements of coal reflectance were taken in essentially monochromatic light, using a G.A.B. interference filter (American distributor - Photovolt Corporation) with maximum transmission at about 530 millimicrons. Figure 1 shows the spectral response of this filter.

DISCUSSION

Variation of Reflectance of Vitrinites as a Function of Experimental Conditions

Many factors in the experimental conditions contribute to variation in reflectance that either masks or exaggerates the inherent variation of the coal substance. In this study most such effects were evaluated by using a standard form of the two-factor form of the analysis of variance (Dixon and Massey, p. 127-139).

It was determined by means of this statistical test that variance of hourly measurements of the same spot was evaluated as being not significant at the 95 percent probability level. Thus it can be assumed that experimental error due to the photometer itself is negligible. It was further determined that if the field diaphragm of the vertical illuminator were stopped down completely and the aperture diaphragm were opened at any position other than completely open, the day-to-day variance was not significant (95 percent level). Variance of reflectance of different spots of the quartz was not significant but was barely so on different spots of the ruby. For this reason the same spots on both ruby and quartz standards were used in all calibration tests.

Numerous tests of significance using the two-factor form of the analysis of variance were used to establish the best (least variance) method of preparing specimens for measurement. With the methods used the variance introduced by repeated polishing and cleaning of the specimens was not significant. Thus it seems justified to assume that the variance within and between coal components as discussed below is real and not a function of experimental error.

Variation of Vitrinites with Rank

The variation of reflectance of vitrinites with percentage of fixed carbon is shown in figure 2. Average reflectance and maximum and minimum values for each sample are shown. These values were determined from the measurement of a variable number of vitrain bands in each polished block. Five points were measured in each band. The progressive increase in reflectance with higher ranks of coal is the most prominent feature of the curve. Even though many points representative of coals of intermediate ranks were not determined, there is an approximate fit to a continuous curve (shown by the dashed line). There is no a priori reason

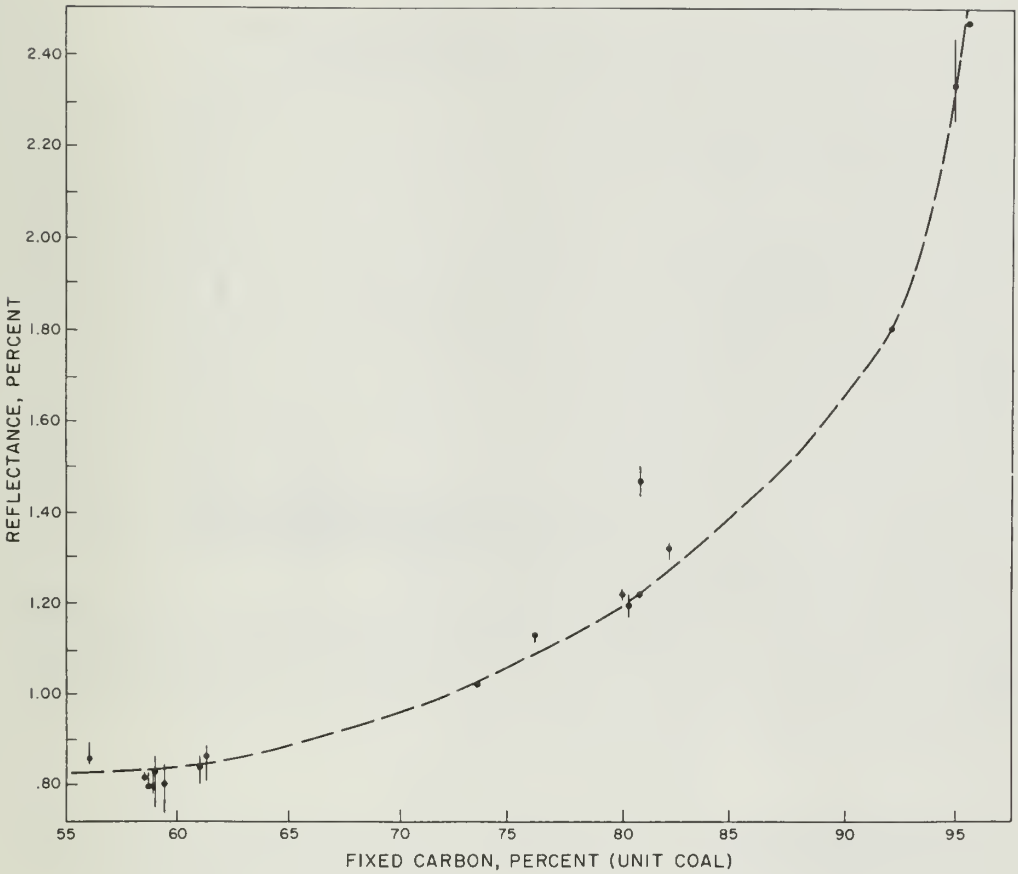

Fig. 2. - Variation of reflectance with rank.

to infer a discontinuous, stepwise increase in reflectance from the results. For a complete discussion of increase in reflectance with rank, see Broadbent and Shaw (1955).

Variation Within and Between Vitrain Bands

Variation of vitrinites within vitrain bands is, in most coals, negligibly small. Horizontal and vertical variation within a vitrain band was investigated in three high-volatile A coals (High Splint, Pewee, Jellico) and the results for all three were similar. Horizontal (parallel to banding) variation was so small as to be negligible. Vertical (normal to bedding) variation was not significant (95 percent level) as evaluated by the two-factor form of the analysis of variance, but did show minor fluctuations which may be real. In each case the reflectance is uniform across most of the band but there are slight decreases in values in 50-100μ wide marginal portions of the band. If this effect is real and not due to polishing relief, it may represent the result of some chemical exchange between adjacent bands of different composition. An alternative, but less likely, explanation is that the reflectance difference is due to slight marginal alterations inherited from the plant materials at the time of peat accumulation.

Variation between vitrain bands in a block of coal is significant in many samples. This variation shows systematic correlation neither with thickness of the band nor with the nature of the coal components on either side of the band. Neither does the variation of reflectance of the vitrain show any definite relation to the structure of the band, that is, the presence or absence of well-preserved cellular structure in the band. It is obvious, therefore, that not all vitrain bands have precisely the same chemical composition, the various slightly different compositions probably being inherited from plant materials of slightly different nature.

In two samples of high-volatile A coals (River Gem, High Splint) the variance between vitrain bands within a small block of coal (approximately 1 inch cube) is greater than the variance between the average values for vitrain bands in different blocks of coal taken from the same place in the mine. In this test 5 vitrain bands in each of 3 blocks of the same coal were compared. This would indicate that the average composition of the vitrain in a coal bed is fairly uniform over very small areas if the differences between bands within a block is subtracted. Perhaps this reflects the character of the original peat that was a highly varied assemblage of plant remains at any point but whose average composition was much the same over small areas in the swamp.

The lower rank coals studied in this investigation (high-volatile A) showed more variation between vitrain bands in the same block than did the higher rank coals. This is probably a consequence of the gradual blurring of compositional differences within and between various coal components in the higher stages of coal metamorphism pointed out by Schopf (1948).

Variation of Components

A comparison of reflectance values of vitrinites, fusinites, semi-fusinites, and micrinites within blocks of coal were made of three coals (River Gem, High Splint, and Illinois No. 5). In this series of measurements, essentially all of the components other than resins, waxes, spore exines, and cuticle (resinite, sporinite, cutinite) were evaluated. Thus the series included all of the coal components called "intermediates" by Seyler (1943, p. 136).

The measurement of reflectance of the "intermediate" components is difficult because of problems of identification under the microscope. The most difficult problem is the distinction between semi-fusinite and fusinite, for one of the primary bases for identification _is_ reflectance, and it would be circular reasoning to attempt to correlate the two objectively when one is dependent on the other.

For transitional components, in the absence of any generally accepted arbitrary reflectance boundary, identification must be based on the structure and appearance of the component. Using structural and textural characteristics, one can readily classify the majority of vitrinites, fusinites, micrinites, and semi-fusinites. But there is a significant proportion of intermediates, up to 15 percent in some coals, which almost defy classification as, for example, either semi-fusinite or fusinite. Thus in figures 3, 4, 5, and 6, where reflectance frequencies are bracketed by component, the classifications must be interpreted with caution.

Figure 3 shows the frequencies of reflectances of vitrinites, fusinites, and intermediates in the River Gem, a high-volatile A coal. In this coal the highest

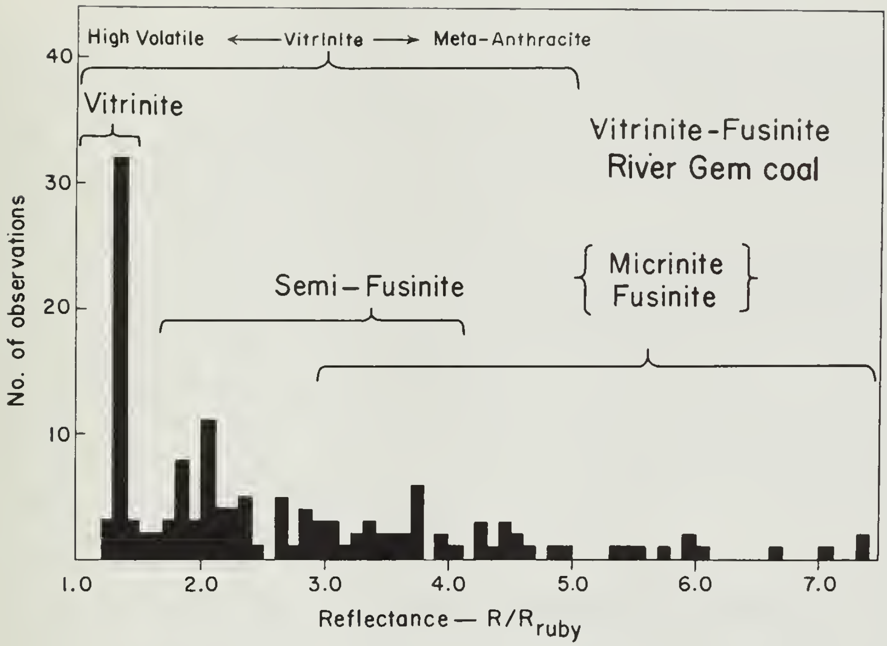

Fig. 3. - Reflectance of components of River Gem coal.

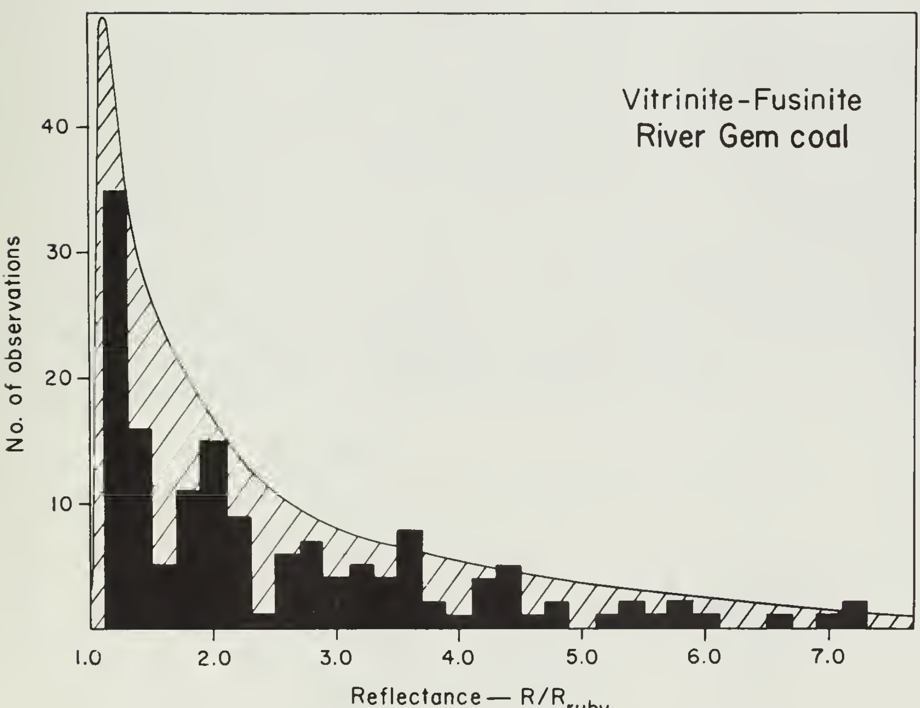

Fig. 4. - Reflectance of components of River Gem coal with superposed gamma distribution curve.

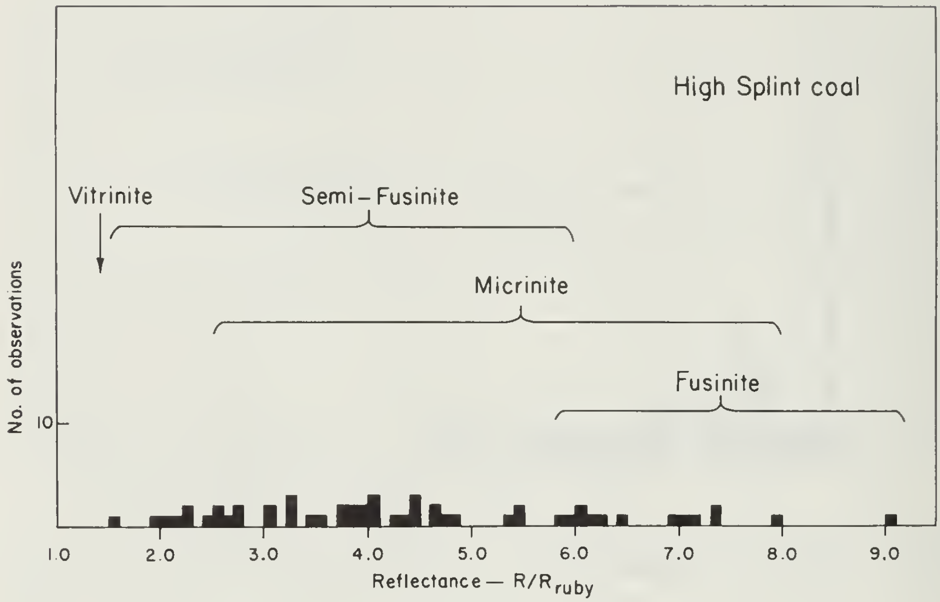

Fig. 5. - Reflectance of components of High Splint coal.

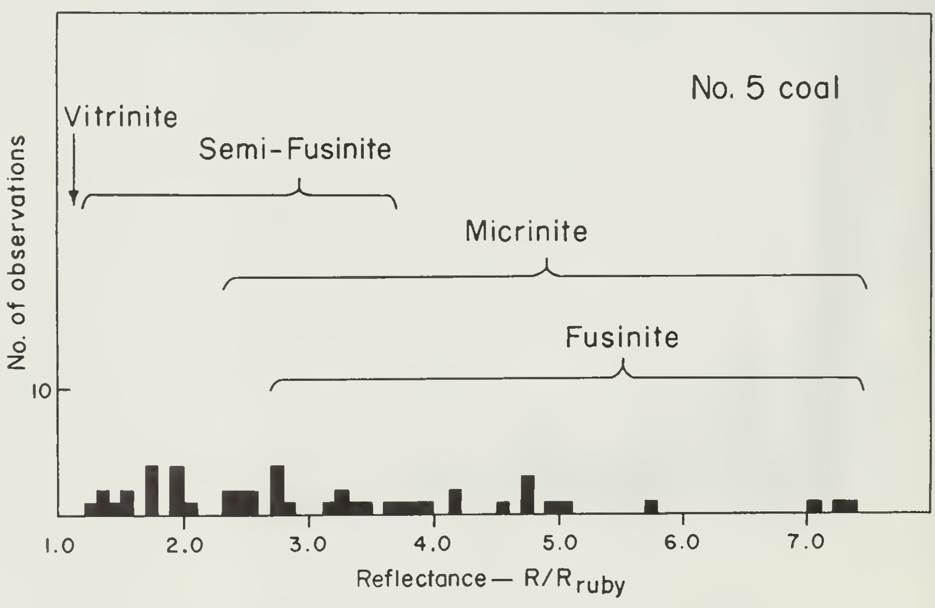

Fig. 6. - Reflectance of components of Illinois No. 5 coal.

reflectant fusinites have a reflecting power about seven times that of the average vitrinite, and almost every value between is represented. In this, as in most other coals, almost all of the vitrinites cluster around the same value, but the other components show a wide range of variation.

Figure 4 shows the same data, recast into larger classes of reflectance. Superimposed on this histogram is a theoretical gamma distribution curve which approximates the histogram in general character.

Figures 5 and 6 show much the same data for two other coals, but with the difference that only the average vitrinite reflectance is indicated and that the histograms do not represent true frequencies but simply distributions of various reflectances to show that the range of values is much the same.

The most significant feature of these histograms is that there is a more or less continuous distribution of reflectances from vitrinites to fusinites and that any petrographic identifications based on reflectivity will have to be made using somewhat arbitrary divisions. The only suggestion of a discontinuity is between the very highest reflectant fusains and all of the other components. This may not be a real discontinuity, however, and perhaps is just an expression of the very low frequency of such highly reflectant materials.

These conclusions point to an obvious need of future research in the combination of petrographic and reflectance studies. At present the distinction between semi-fusinite and fusinite or between vitrinite and semi-fusinite is based on dual criteria - structure and form, and reflectance. More intensive work on the transitional components is needed in order to establish any natural boundaries based on either criteria and to see whether identifications based on structure and reflectance can be made completely internally consistent.

Another fertile field for future research lies in the systematic study of reflectance variations within coals of the same rank in any particular area, within the same coal in a mine, in a small local area, and in a large region such as a depositional basin. A useful tool for the study of these kinds of variability is the hierarchical case of the analysis of variance (Dixon and Massey, 1951; Hald, 1952), with which the variability on different levels of area, for example, can be evaluated simultaneously and the variance within each area can be estimated.

Based on such studies, a practically useful classification of coal using reflectance criteria may be possible. Such a classification will be rapidly and easily performed and also should have the advantage of being based on non-empirical analyses of one of the fundamental physical properties of the homogeneous petrographic components that make up the coal substance.

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Illinois State Geological Survey Circular 241
11 p., 6 figs., 1 table, 1957

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CIRCULAR 241

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