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**THE MEASUREMENT OF REFLECTANCE OF COAL MACERALS—ITS
AUTOMATION AND SIGNIFICANCE**

Technical Report 10

By
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MASTER

U. S. DEPARTMENT OF ENERGY

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ABSTRACT

One of the prime objectives of The Pennsylvania State University's DOE program is the characterization of 1,300 samples of U.S. coals. Reflectance determination plays a major role in meeting this objective, because it is used as an accurate rank index both to select coals for testing and to investigate property-behavior interrelationships using the Penn State/DOE Coal Data Base.

The reflectances of coal macerals are related to their refractive and absorptive indices by the Beer equation; the refractive index of the medium and the wavelength of light need to be specified. Determinations usually are made in immersion oil at 546 nm. The properties of vitrinite make it the most suitable maceral for these measurements, but the variety of vitrinite selected may depend upon the intended application. Vitrinite reflectance is considered to be dependent upon the degree of aromatization of its structural units.

Although some of the earlier investigators believed that the reflectance of coals increased in a discontinuous, stepped manner as their rank increased, the bulk of assembled data suggests that the change is continuous. However, some recent results indicate that there may be more than one coalification track for coals with different geological histories, resulting in displacement from the general trend.

Several techniques have been employed to determine the optical properties of coal constituents. The usual comparative method involves the use of a microscope photometer to compare the intensity of light reflected by particles within a polished pellet of coal to that of a glass or synthetic mineral standard. Because coal is anisotropic it is common to measure either maximum reflectance in polarized light or random reflectance in non-polarized light. Various equipment modifications and accessories have been used to improve the ease of measuring maximum reflectance.

Reflectograms produced by fully automated reflectance microscopy (ARM or Rapid Scan) can be interpreted to obtain measurements of vitrinite maximum reflectance and maceral composition. Earlier attempts had been limited severely by the technology available, but current work at The Pennsylvania State University has the benefit of a modern minicomputer facility. The very rapid data acquisition rate achievable with a minicomputer makes it an excellent tool to couple with a microscope photometer. The difference in reflectance between the organic components of coal and pyrite enables pyrite chord-size distributions to be measured by summing consecutive readings on pyrite during continuous scanning. The system gives reproducible results, and, because it is capable of simultaneously quantifying rank, petrography, and pyrite size and amount, it holds promise as a means of coal quality control. Further work, aimed at gaining resolution from the use of a pulsed step scanning stage, is in progress.

Vitrinite reflectance provides a rapid, accurate measurement of coal rank requiring only a small amount of material. Consequently several systems of classifying coal and of establishing a petrographic basis for understanding coal composition and behavior are based on reflectance. Reflectance limits for ASTM rank classes have been specified, based on results for 140 Penn State/DOE coals.

A principal application of reflectance data is in the prediction of the coking properties of coal. Other uses are as a geothermometer, as a means of estimating the petroleum-bearing potential of sedimentary sequences, and physical constitution analysis to derive information on the chemical structure of coal.

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I. INTRODUCTION

Reflectance measurement has been accepted as a rapid and accurate index of coal rank, being largely uninfluenced by the petrographic variation which can affect the results of many of the chemical tests used for the same purpose. One of its principal advantages is that it can be made on extremely small amounts of material.

A major objective of The Pennsylvania State University's DOE program is the detailed characterization of 1300 samples of U.S. coals. The determination of reflectance has a prominent role in this characterization, because it is used as a rank index to select samples for testing, and for investigating the interrelationships between compositional properties and technological behavior using the Penn State/DOE Coal Data Base. Reflectance is also the means of discrimination used in the Rapid Scan system of automated microscopy being undertaken as part of the program; it is anticipated that this facet of the research will lead to the development of a system suitable for simultaneous routine quality control of the rank or mix of ranks and of the maceral composition of a coal or coal blend, and of the amount and size distribution of the pyrite content.

Now that it is becoming a more commonly employed procedure, reflectance determination is being used in the U.S. as a means of studying the causes of coalification, as a geothermometer for measuring paleotemperatures, in determining the likelihood of petroleum occurrence in sedimentary rocks, and for predicting the coking behavior of coals.

Consequently, it seems appropriate to concern this report with the theory, of means and methods of reflectance determination, to review some of the results obtained and conclusions drawn from them about the nature of coal, and to present some of the applications of these measurements, including automated reflectance microscopy.

II. OPTICAL PROPERTIES OF VITRINITE AND OTHER MACERALS

A. Vitrinite Reflectance and Its Relationship to Refractive and Absorption Indices

Reflectance may be defined as the proportion of normally incident light that is reflected by a plane, polished surface of the substance under consideration. It is usually expressed as a percentage, so that an oil reflectance of 1.00% for vitrinite indicates that only that proportion of the light incident on the coal surface is reflected under the conditions of measurement. The term "reflectivity" has been used synonymously with reflectance, although some authors have restricted use of the latter to the specific conditions (oil immersion, green light) which normally apply for measurements on coal.

For most purposes it is the reflectance of the vitrinite component that is determined, for the following reasons:

- (i) Vitrinite is the preponderant maceral in most coals.
- (ii) Vitrinite often appears homogeneous under the microscope.
- (iii) Particles of vitrinite are usually large enough to permit measurements to be made easily.
- (iv) In the application of petrographic techniques to the industrial uses of coal, particularly in carbonization, interest is focussed on the behavior of vitrinite which, is the maceral principally responsible for the plastic and agglutinating properties of coal.

The purpose of the reflectance determination influences the selection of the type of vitrinite on which the measurements are taken. Where reflectance is being used as an index of rank or degree of metamorphism, it is better to make determinations on as restricted a range of vitrinitic materials as possible. The Handbook of the International Committee for Coal

Petrology (ICCP, 1971) states that for this purpose the reflectance of the homogeneous collinite which generally occurs in wide layers (telocollinite) normally should be measured, rather than that of the finely attrital desmocollinite or other types of vitrinite (Figure 1a). In the case of brown coals, lignites, and some subbituminous coals, an even narrower restriction may be necessary; McCartney and Teichmüller (1972) suggest that the most suitable of the low-rank huminite macerals for reflectance measurement is the more highly reflecting type of ulminite, which, although dominant in many U.S. lignites, is only one of the several possible vitrinite precursors.

Where it is the intention to characterize all of the vitrinite, as for example where the potential behavior of the whole of the coal in a coking process is being assessed (see Section VIII), then a statistical coverage of all of the vitrinite present is necessary. In the method of coke-strength prediction developed by Schapiro *et al.* (1961) such a procedure is in fact followed. However, Koch (1970) and others have observed that it has yet to be proved that different vitrinites of the same reflectance from mixed-frequency distributions have the same technological behavior. The method used by Bethlehem Steel Company for predicting coke properties (see Benedict *et al.*, 1968) requires that separate reflectance values be obtained for "pseudovitrinite" (Section VIII) and "reactive vitrinite." The higher reflecting pseudovitrinite (Figure 1b) appears to be equivalent in part to telocollinite and to vitrinite A of Brown *et al.* (1964). Vitrinites A and B are respectively the higher reflecting vitrinite which occurs in thicker vitrain bands, and the lower reflecting vitrinite of layers of mixed macerals; they normally have reflectance distributions which overlap, as do those of reactive vitrinite and pseudovitrinite (Koch, 1970). Use of the reflectance of "reactive vitrinite" as an index of rank can give lower rank values

Figure 1. MICROSCOPIC APPEARANCE OF VARIETIES OF VITRINITE IN REFLECTED LIGHT UNDER OIL IMMERSION

- (a) The light-grey homogeneous band in the upper part of the photomicrograph is of telocollinite (t), bounded by thin dark-grey layers of cutinite. The lower part of the field is of attrital coal, containing fragments of desmocollinite (d) which are somewhat lower in reflectance than the telocollinite.
- (b) The particle of pseudovitrinite (p) in the upper center of the photomicrograph has a higher reflectance than the vitrinite contained in the other large particles.

than use of either a coverage of all vitrinite types or a selection of readings from only particles of telocollinite. It is possible to measure the reflectances of the different types of vitrinite separately and combine the results mathematically according to the proportion of each that is present. In this way any of the reflectance values or ranges can be utilized.

The reflectance in oil of an absorbing medium such as vitrinite is considered to be given by Beer's equation (Eq. (1)):

$$R_o = \frac{(n - n_o)^2 + n^2 \kappa^2}{(n + n_o)^2 + n^2 \kappa^2} \dots \dots \dots (1)$$

where n and κ are respectively the refractive and absorption indices of the vitrinite, and n_o is the refractive index of the immersion oil (normally taken as 1.518 at the standard wavelength of 546 nm).

Ideally, n and κ should have been calculated previously, requiring that reflectance be determined separately in two different media (air and oil are convenient), using Eqs. (2) and (3), which are based on Eq. (1).

$$n = \frac{\frac{1}{2}(n_o^2 - 1)}{\frac{n_o(1 + R_o)}{(1 - R_o)} - \frac{(1 + R_a)}{(1 - R_a)}} \dots \dots \dots (2)$$

$$\kappa^2 = \frac{R_a (n + 1)^2 - (n - 1)^2}{n^2 (1 - R_a)} \dots \dots \dots (3)$$

where R_a and R_o are respectively the measured reflectances of the vitrinite in air and oil, and n_o is the refractive index of the oil at the temperature at which the determination is made. In Eqs. (1)-(3) reflectances are expressed as fractions, not percentages.

In practice, it is acceptable to determine the reflectance in oil of vitrinite by direct comparison with a reflectance standard and without recourse to the above equations, by assuming the refractive index of the immersion oil.

Figure 2 illustrates in the manner of Huntjens and van Krevelen (1954) how the reflectance of vitrinite is dependent upon both the refractive and absorption indices. In lower rank coals, the refractive index has the dominant influence, whereas once the refractive index has reached about 2.0 it shows little further increase, and the absorption index becomes the principal controlling factor. The maximum refractive index of graphite was determined from reflectance measurements to be about 2.0 by Huntjens and van Krevelen (*ibid.*), and 2.15 by McCartney and Ergun (1958).

B. Anisotropy of Vitrinite

Vitrinite usually behaves as a uniaxial negative substance, with the optic axis of the indicatrix normal to the bedding plane (Figure 3). Consequently, a surface that is perpendicular to the bedding displays a maximum refractive index, and thus reflectance, when the incident light is polarized in a direction parallel to the bedding, and a minimum reflectance when the light is polarized perpendicular to the bedding; the difference between the maximum and minimum reflectances is termed bireflectance. A surface parallel to the bedding displays maximum reflectance in all directions. On an oblique surface of coal, maximum reflectance is displayed parallel to the bedding, and intermediate values (apparent minima) between maximum and minimum are shown at right angles to that direction. Therefore a maximum reflectance value can always be obtained with a reflectometer by rotating any coal particle under the microscope until the bedding direction is parallel to the plane of vibration of the polarized incident light. Chiefly for this reason, many petrologists have preferred to use maximum reflectance as an indicator of

Figure 2. THE RELATIONSHIP BETWEEN THE REFLECTANCES OF COALS AND THEIR REFRACTIVE AND ABSORPTION INDICES

Figure 3. THE ANISOTROPIC CHARACTER OF VITRINITE

The range in reflectance in polarized light is dependent upon the orientation of the polished surface with respect to the bedding. R_{ω} and R_{ϵ} are the maximum and minimum reflectances, respectively. $R_{\epsilon'}$, an apparent minimum reflectance, is intermediate between R_{ω} and R_{ϵ} .

(Modified from Davis, 1975)

rank. In the United States, this procedure is followed almost exclusively, and is required in the method of the American Society for Testing and Materials (ASTM, 1967b).

As just discussed, individual readings on several randomly oriented particles might vary from true maximum to true minimum in polarized light. The mean of a number of such readings is the mean random or average reflectance. In non-polarized light the individual random-reflectance value of a vitrinite particle is assumed to be independent of the orientation of its surface but dependent upon the particular surface presented. The random reflectance displayed by each particle should be the mean of the maximum and apparent minimum reflectances, so that individual values obtained on a particulate sample could vary between maximum and the mean of maximum and true minimum reflectances. The range of values therefore is much less than it is when polarized light is employed. Partly for this reason, mean random or average reflectance usually is determined in non-polarized light.

According to Hevia and Virgos (1977), the statistical mean value for random reflectance is related to the true maximum and minimum reflectances as follows:

$$\bar{R}_e = \frac{2R_a + R_c}{3} \quad \dots \dots \dots (4)$$

where $\bar{R}_e$ is the random reflectance and R_a and R_c are the maximum and true minimum reflectances in the principal directions of the vitrinite.

The advantages of the use of maximum rather than random reflectance include the following:

- (i) Although there are variations due to the inhomogeneity of vitrinite,

maximum reflectance is a natural parameter of the degree of a coal's metamorphism.

- (ii) There is much less standard deviation or scatter for maximum reflectance. That of random reflectance is, in turn, less than that of minimum reflectance (de Vries *et al.*, 1968; Pitt, 1975).
 - (iii) As a consequence of (ii), one may more easily discriminate between individual coals in a blend using maximum reflectance (see de Vries *et al.*, 1978).
 - (iv) Because light from the source is partially polarized, the condition for random reflectance measurement in "non-polarized light" strictly may not apply.
 - (v) Two of the major methods of coke-strength prediction used in the United States are based upon determination of maximum reflectance.
- On the other hand, points favoring the use of random reflectance are:
- (i) The time-consuming rotation of the microscope stage is avoided.
 - (ii) The measurement avoids errors due to lack of centering and change in focus on rotation.
 - (iii) Reflectance measurements on the dispersed organic matter of sediments are used particularly in studies of the degree of metamorphism of rocks with petroleum-bearing potential. Readings on particles which are sometimes smaller than $5\text{ }\mu\text{m}$ may preclude rotation of the microscope stage. In any case, such work does not require absolute values, only reference to a suitable empirical scale.
 - (iv) Removal of the polarizer provides more light and the possibility of achieving higher signal/noise ratios.
 - (v) In some practical applications, the difference in resolving power between maximum and random reflectance measurements is not sufficiently

large to be of importance (Pitt, 1975).

- (vi) According to Hevia and Virgos (1977), the existence of some bi-axial anthracites invalidates the use of maximum reflectance because this value varies between the extremes representing two of the principal directions of refractive index. However, Cook *et al.* (1972) maintain that maximum reflectance from particulate blocks of such high-rank coals is nevertheless the most useful measurement of rank.

C. Reflectance of Other Macerals

Few data have been published on the reflectance of macerals other than vitrinite. Figure 4, after Dormans *et al.* (1957) shows the relative reflectances of vitrinites, exinite and micrinite at different carbon contents of the vitrinites. The decrease in contrast in appearance of these macerals which is experienced at higher levels of rank, and the variation in rate of change of reflectance with rank are apparent in this figure.

Fusinite is generally the highest reflecting maceral. For a Queensland Permian coal in which the mean maximum reflectance of the vitrinite is 0.56%, those of semifusinite and fusinite have been found to be 1.26 and 2.32%, respectively. For an Illinois coal with vitrinite reflectance of 0.66%, the semifusinite and fusinite values were 2.50 and 4.82%, respectively. A Pennsylvania anthracite contains fusinite with a maximum reflectance of 4.65%, approximately the same as that of the vitrinite. Alpern and Lemos de Sousa (1970) have reported a peranthracite containing fusinite with a maximum reflectance of 5.20%, lower than that of the vitrinite which was 7.30%.

Reflectance ranges for other macerals, including those of brown coals, are given in ICCP (1971) and Stach *et al.* (1975).

Figure 4. THE REFLECTANCES OF THREE MACERALS AT DIFFERENT LEVELS OF RANK
(from Dormans et al., 1957)

D. Reflectance at Different Wavelengths

In general, coal petrologists have determined the optical properties of coal constituents at about either 530 nm or 546 nm. However, results have been quoted at other wavelengths, and many authors fail to state the particular wavelength, if any, at which their readings were taken. Today, the ASTM and ICCP methods use the wavelength corresponding to the mercury line within the green region (546 nm).

Very little work has been published on the variation of optical properties of coal macerals with wavelength, although the small amount of data acquired has led to some interesting conclusions concerning the structure of coal.

As a result of measurements conducted with a spectrophotometer, Friedel (1959) claimed that vitrain with 84% carbon displays no marked change in reflectance or refractive and absorption index between 200 and 600 nm. Such changes would be expected in regions of strong absorption, so he concluded that only small amounts of polynuclear condensed aromatics, which show pronounced absorption bands in the ultraviolet, can be present in coal. However, Gilbert (1960) noted that Friedel's data seemed to point to a decrease in reflectance and refractive index for the lower wavelengths, together with an increase in the absorption index. He concluded initially from his own experiments, using a macroscopic photometric method, that there was no evidence for the presence to any marked extent of simple aromatic substances in vitrains, but that complexity in the nature of any aromatics present increased with rank. This last conclusion was based on the smoothing out of the reflectance curves of higher rank coals. Figure 5 shows the contrast which Gilbert found between the reflectance spectra of bituminous coals and anthracites. He was able to show later (1962) that

Figure 5. VARIATION OF THE REFLECTANCES OF VITRAINS WITH WAVELENGTH
 (1) 85% Carbon; (2) 87.6% Carbon; (5) 94% Carbon; (6) 93.9% Carbon
 (from Gilbert, 1960)

there was indeed evidence of strong absorption in the ultraviolet spectrum, which could be interpreted as being due to the presence of aromatic substances.

Cannon and George (1944) have published refractive-index dispersion curves for four coals, all rising towards the shorter wavelengths of the visible spectrum. The slopes of the curves for sections of the two high-rank coals polished parallel to the bedding were steeper than those of surfaces perpendicular to the bedding. Whereas Gilbert (1962) and McCartney *et al.* (1965) were able to find different shaped curves for the refractive index dispersion of anthracites, Cannon and George's samples were not sufficiently high in rank to record this distinction.

Jones (1961) has measured the reflectance dispersions and calculated the dispersions of refractive and absorption indices for a number of vitrinites, and his results, determined by a microscopic photoelectric method, agree with the corresponding results of Gilbert. He showed that there was a pronounced difference in the shape of dispersion curves for absorption and refractive indices of vitrinites below and above a refractive index value of 1.80 at 532 nm. Such a change was not as pronounced for air reflectance dispersions, but was nevertheless discernible.

Air reflectance dispersion curves obtained by the author (Davis, 1965) demonstrate a definite change in shape for coals with reflectances greater than 12% (Figure 6a). The curves for the anthracitic vitrinites have overall dispersions which are much less than for the lower rank materials; in fact the upper curve shows a decrease in reflectance for progressively lower wavelengths over most of its range, in support of Gilbert's results (1960). The transmittances of the 410 and 440 nm filters used by the author were

relatively low, and the readings at these two wavelengths were not as accurate as at higher wavelengths.

Figure 6b shows oil-reflectance dispersion curves for four groups of vitrinites; the flattening of the curves for the anthracites that was noted in the air dispersion curves is not as clear in the case of these spectra. Jones (1961) also observed that the oil dispersion curves do not respond in so marked a manner as the air spectra. Ergun and McCartney (1960), and Hevia (1974), however, have reported reversals in the slopes of the oil-reflectance spectra for a Pennsylvania anthracite (95% carbon; dry, ash-free) and a Spanish meta-anthracite, respectively; in these cases the vitrinite reflectance clearly increased with increasing wavelength.

The refractive index dispersion curves in Figure 6c support other reports of a reversal in the trend of such curves for the higher rank vitrinites following a decrease in dispersion with rank through the bituminous range. This reversal is very apparent in these results in spite of an anomalous value occurring at 482 nm in the dispersion curve for Vm/35a. The dispersion curves in Figure 6 are for single vitrains and groups of vitrains of very similar rank.

Murchison and Jones (1964) have reported that resinite shows a pattern of increasing reflectance with decreasing wavelength, similar to that of the vitrinite from the same bituminous coals, although the increase is less steep in the case of the resinites, especially in the lower rank coals. Hevia (1974) has published spectra for macerals from a single coal; the slopes of the curves for vitrinite, resinite, and sporinite are very similar. However, fusinite, semifusinite, and macrinite have curves whose slopes decrease progressively as the mean reflectance of the maceral increases. Generally the slopes of the curves for inertinite group macerals are less than those of vitrinite of the

Figure 6. VARIATION OF THE OPTICAL PROPERTIES OF VITRINITES WITH WAVELENGTH

(a) Air Reflectance; (b) Oil Reflectance; (c) Refractive Index

same level of reflectance; they also tend to be somewhat more irregular than those of vitrinite. Further, the spectra of inertinite of higher reflectance apparently show minima in the range 480-550 nm.

E. Review of Previous Results and Conclusions

A relationship between the reflectance and rank (volatile matter) of coal was reported first by Hoffmann and Jenkner (1932). Seyler (1943) obtained a number of results with a Berek microphotometer, from which he deduced that there was a discontinuous change in reflectance in oil with coal rank (Figure 7). In all, Seyler established ten components and claimed that the reflectances of the wood-derived constituents of coal could be assigned to one of these components, any variation from the component value being caused by experimental error. These components had reflectances in oil of 0.26, 0.34, 0.51, 0.67, 0.92, 1.26, 1.64, 2.23, 3.11, and 4.17%, there being an approximate relationship among these values which could be expressed as:

$$R_o = 0.26 \times 1.363^{Nr} \dots \dots \dots (5)$$

where R_o is the reflectance in oil and Nr is an integer,
1 through 9.

Some investigators claimed to have confirmed Seyler's stepwise series of coal-reflectance classes (Sherlock, 1951; Mukherjee, 1952; Shaw, 1953; Huntjens and van Krevelan, 1954). Shaw, employing a microscope-photomultiplier combination, found some confirmation of Seyler's results in that discontinuities in the trends of reflectance and refractive index were shown at carbon contents of approximately 80 and 92%.

Figure 7. STEPWISE VARIATION IN REFLECTANCE OF VITRINITES WITH CARBON CONTENT
(from Seyler, 1944)

Huntjens and van Krevelen (1954), while agreeing that "centres of gravity" occurred around Seyler's values, could find no evidence to support the existence of clearly defined components at 0.92 and 1.26% reflectance. These investigators used the subjective Berek method, although they found it gave comparable results to an objective photographic method.

The bulk of the reflectance data which have been assembled points to the conclusion that there is no stepwise or discontinuous change of reflectance with rank. Dahme and Mackowsky (1951) were the first to contradict Seyler's conclusion, having used a selenium cell to obtain values of reflectance in air for which they claimed an accuracy of $\pm 0.05\%$. McCartney's earliest results (1952) were obtained using a Berek microphotometer and also failed to confirm Seyler's steps.

Broadbent and Shaw (1955) applied a lumped-variance test to much of the previous data, as well as to their own which was obtained using a photomultiplier, and indicated that a continuous variation of reflectance was indeed possible. Seyler and Chandra (1955) managed to develop a series of "bathric" (stepped) curves by joining the experimental plots of Broadbent and Shaw's results. The steps occurred at about 82.0, 84.5, 87.5, 89.3 and 91.8% carbon, three coinciding approximately with Seyler's steps.

As more data are accumulating, the need to re-examine critically the continuity of the variation in reflectance with rank may have arisen. Thompson and Benedict (1974) have presented a relationship between reflectance and volatile matter for selected American coals (Figure 8) which suggests to them that there are discontinuities representing fundamental changes in the character of coal; they have also observed that the change in slope of their curve may be related to important differences in coking behavior.

Figure 8. DISCONTINUOUS RELATIONSHIP BETWEEN REFLECTANCE OF REACTIVE VITRINITE AND COAL VOLATILE MATTER
(Modified from Thompson and Benedict, 1974)

They refer to discontinuities at about the 0.7, 1.0, and 1.35% reflectance levels of "reactive vitrinite" (i.e. vitrinite other than "pseudovitrinite").

Benedict (1977) has observed that the offset upper left sector of the curve in Figure 8 represents coals from the Midwest of the United States (principally Illinois, Indiana, and west Kentucky). Waddell (1978) also has observed that vitrinites from the Eastern region of the Interior coal province fall on the low-reflectance side of the reflectance-carbon content curve.

Davis (1965, 1971) has reported that vitrinites from Northumberland, United Kingdom, have lower reflectances than other Carboniferous and Permian vitrinites of equivalent carbon content (Figure 9). The solid line in the figure represents a compilation by van Krevelen (1961) of reflectance determinations on Carboniferous coals. Davis found that some Permian vitrinites fell close to this trend, as did his results for British vitrinites with greater than 86% carbon. The British coals of lower carbon content were collected from Northumberland and north Durham, and the results for these are represented in the figure by a broken line. The observation of the relatively low reflectances of Northumberland coals is supported by the data of Jones (1961). Northumberland vitrinites have a strongly mottled appearance due to darkening of the coalified secondary cell-wall tissues; this fact and the generally lower hydrogen values of these vitrinites do not support the suggestion of some authors that the mottling is due to incorporation of resinous material. The threshold between the higher and lower reflectance groups occurs at about 85-86% carbon or about 0.8-0.9% reflectance. Thompson and Benedict's discontinuity (for "reactive vitrinite") occurs around a 0.7% reflectance level, and Seyler interpreted his data as representing a reflectance step from an average of 0.67 to 0.92% at about 85% carbon. Thompson and Benedict have suggested that at certain rank levels, a coalification

Figure 9. VARIATION IN MAXIMUM REFLECTANCE WITH CARBON CONTENT, ILLUSTRATING THE LOWER VALUES OBTAINED FOR NORTHUMBERLAND VITRINITES

jump or mutation similar to that reported by Stach and Michels (1955) for the coalification of exinites may occur. Alternatively, the data may reflect differing geological histories of coals from different basins, and the existence of more than one vitrinite coalification track.

McCartney and Teichmüller (1972) postulate that vitrinite reflectance is fundamentally dependent upon the degree of aromatization and condensation of the structural units. Refractive index, they observe, is a function of atomic density and increases with aromatization. Absorption depends upon the number of delocalized electrons. The mobility of these electrons increases with the size of the aromatic complex and degree of condensation.

There is a good general relationship between reflectance and aromaticity. In Figure 10, aromaticity was calculated by the graphical densimetric method of physical constitution analysis; certain coals are displaced mainly because of the effect of low hydrogen values on the calculation of aromaticity (Davis, 1965).

Reflectance is regarded by many as perhaps the best single parameter of coal rank (McCartney and Teichmüller, 1975). The curves in Figure 11 shows how reflectance varies with carbon content in plots of data reported by several workers.

Cannon and George (1944) and Dahme and Mackowsky (1951) demonstrated that a section of vitrinite parallel to the stratification is effectively isotropic, and that one perpendicular to the stratification displays a maximum refractive index and reflectance when the electric vector of the light wave is parallel to the stratification, a minimum when it is normal to the stratification. This general orientation of the anisotropy of vitrinite is due to the influence of load pressure. However Jones *et al.* (1973) describe the anisotropy of vitrinite which parallels the grain of the original wood in vertical tree barks.

Figure 10. VARIATION IN REFLECTANCE WITH AROMATICITY (GRAPHICAL DENSIMETRIC METHOD)

Figure 11. THE RELATIONSHIP BETWEEN MAXIMUM AND MINIMUM REFLECTANCE OF VITRINITES AND CARBON CONTENT
Data from Huntjens and van Krevelen (1954); Broadbent and Shaw (1955); Murchison (1958); and Davis (1965)

Dahme and Mackowsky (1951) suggested that coal possesses a layer structure due to the presence of stratified laminae of crystallites. For anisotropy to be developed, the crystallites must be brought close together and must lie together with crystallographically identical orientation. The closer packing of the crystallites takes place with coalification upon loss of volatile constituents, while the alignment is due to either the pressure and temperature of coalification or to a pre-orientation of vegetal tissue. At that time, they supposed that the effect of temperature and pressure upon the development of anisotropy is slight, and that it is largely due to pre-orientation.

Wege (1954) believed that load pressure as well as pre-orientation is important in the development of anisotropy. He cited experiments conducted by H. Winter in 1953 as well as the relationship between anisotropy and stratification in support of his theory. In Winter's experiments, artificial coals prepared at low pressures did not show any reflection anisotropy.

Cannon and George (1944), having observed differences in the refractive-index dispersion curves perpendicular and parallel to the stratification, felt that directed geological stress had produced a preferred orientation. They subscribed to the theory that the birefringence of coals is due to orientation of "platelets" of colloidal dimensions, rather than "rodlets" which would produce a uniaxial positive orientation. Their theory required that the platelets be surrounded by a medium of different refractive index, namely the volatile constituents of coal.

Williams (1953) reported that the anisotropy of some high-rank coals had decreased in the vicinity of thrust faulting, and related the formation of anisotropy to tectonic pressure. Petrascheck (1954) has described obliquely oriented anisotropy resulting from fold pressure. Davis (1971)

also has described non-parallel anisotropy for some high-rank vitrinites from the Bowen Basin, Australia. Cook *et al.* (1972) have shown that some anthracites are optically biaxial.

There is some variation in published results as to the rank of coals in which anisotropy first develops. Cannon and George (1944) observed that the onset of anisotropy occurs at about 89.6% carbon, but is not significant until the coal reaches semianthracite rank. van Krevelen's (1961) results indicate that the least mature vitrinite in which anisotropy could develop is about 81% carbon, although his graphs representing determinations of reflectance in oil by a number of workers show that the effect can be observed for coals of less than 80% carbon. Wege's results (1954) show only a slight increase in anisotropy up to a reflectance of 13 to 14% in air (about 93% carbon), after which it increases markedly. Shaw (1953), Broadbent and Shaw (1955), and McCartney and Ergun (1958) found anisotropy throughout the rank range, and Hirsch (cited by McCartney and Ergun, 1958) has given x-ray evidence of order in all of the coals which he examined.

de Vries *et al.* (1968) claim that only above a reflectance of 1.4% is vitrinite clearly anisotropic. The data of Murchison (1958) and Davis (1965) indicate that the British coals which they examined possessed a measurable bireflectance at the lowest rank (about 82% carbon) and that there was progressive increase above this. In studies of some vitrinites from the Bowen Basin, Davis (1971) found values of bireflectance which were lower than those reported for other coals of equivalent carbon content in the range 87 to 91.5%. Figure 11 demonstrates the increasing anisotropy (difference between maximum and minimum reflectance) which occurs with increasing rank.

III. EARLY APPROACHES TO THE MEASUREMENT OF REFLECTANCE

Several different techniques have been employed in attempts to determine the optical properties of coal constituents. Some of the earliest measurements used direct rather than comparative methods. Quirke and McCabe (1938) have described the use of a goniometer table to measure visually the angle of maximum polarization in reflected light of vitrain samples. The tangent of this angle is equal to the refractive index, according to Brewster's Law. Cannon and George (1944) also made use of the Brewster angle to calculate the refractive indices and absorption coefficients of coal constituents at different wavelengths using a spectrometer table and a photo-cell to record the variations in light intensity. van Krevelen (1961) has observed that the Brewster relationship strictly should not be applied to strongly absorbing media such as coal.

Gilbert (1960, 1962) used a photometer to measure the reflectivity of vitrain macroscopically. One method was by comparison against a fused silica standard, and another was by determining the refractive indices and absorption coefficients from the ratios of signals obtained at two angles of incidence, from light polarized with the electric vector both in and perpendicular to the plane of incidence.

Many of the early determinations of reflectance made use of the Berek microphotometer (Figure 12), a microscope attachment which allows the light from the reflecting surface to be visually matched against a comparison beam of polarized light (B), the intensity of which is varied by rotating a nicol. The reflected and comparison beams are observed simultaneously in adjacent halves of the field of view. Variation in the intensity of the comparison beam is proportional to the square of the sine of the rotation angle of

Figure 12. THE PRINCIPLE OF THE BEREK MICROPHOTOMETER

L, Light Source; A, Reflected Beam; B, Comparison Beam; N₂, Nicol;
 N₁, Comparison Nicol; P, Polarizer; G, Neutral Density Filter
 (from Stach and Michels, 1955)

the rotating nicol N_2 . The use of a standard of known reflectance enables the reflectance of an unknown to be calculated from Eq. (6).

$$R_1 = \frac{\sin^2 \alpha_1}{\sin^2 \alpha_2} \cdot R_2 \dots \dots \dots (6)$$

where R_1 and α_1 are the reflectance and angle of rotation for the unknown, and R_2 and α_2 are the values for the standard.

Stach and Michels (1955) and others have used neutral density filters (Figure 12, G) to reduce the intensity of the comparison beam so that larger rotation of the comparison nicol is required, resulting in a more sensitive and accurate measurement.

The subjectivity of the reference-beam method just described led to the incorporation of a single photocell (Stach, 1956; Stach and Michels, 1955); a rapidly rotating sector allowed the two light beams to fall alternatively upon the cell. Adjustment of the polar to obtain a zero galvanometer deflection enabled an objective measurement to be made.

Because of the possibility of variation in the power supply to the photomultiplier and light source, Jones (1962) modified a system developed by Fenton *et al.* (1958). The incident light beam was passed through a rotating polar, and the sample and comparison beams, which were 180° out of phase because of the introduction of a ring polar into the comparison beam, were matched on an oscilloscope using the Berek matching principle.

The main microscopic procedure that has been employed for measuring the optical properties of coal macerals has been the single-beam comparative method in which the intensities of light reflected by the coal and a standard of known reflectance are recorded successively. Earlier instruments made

use of various photocells, but these were superceded by photomultipliers. A fuller description of this type of instrument will be given in Section IV.

Other comparative techniques for measuring reflectance have been attempted, but have not received a great deal of application. These include photographic methods, both in association with the Berek microphotometer (Huntjens and van Krevelen, 1954), and independently (Holubar and Krejci, 1957), and a continuous grey wedge developed by Stach (1958) which can be matched against the coal components under a comparison microscope.

IV. MICROPHOTOMETRIC APPARATUS AND ACCESSORIES

Although standard methods for the measurement of reflectance have been established, these generally serve as guidelines only; there is much latitude in the type of equipment and techniques that are employed.

Microscope photometers (Figure 13) have three principal components; a reflected-light microscope with a stabilized light source; a photometric device, usually a photomultiplier; and a display and recording system. Essential accessories are a monochromatic filter, an aperture to limit the size of the area under measurement, and a set of standards of known reflectance. An example of a commercial microscope photometer is shown in Figure 14.

A. Microscope System

A suitable polarizing microscope, equipped for reflected-light examination has the following components and accessories:

1. Light Source

High-power lamps should be used to obtain a satisfactory level of intensity from the low-reflecting coal macerals. Tungsten-filament lamps are adequate, but quartz-halogen lamps are now in more general use. A common rating for these lamps is 12V, 100W.

Figure 13. SCHEMATIC DIAGRAM OF THE OPTICAL COMPONENTS OF A MICROSCOPE PHOTOMETER

Figure 14. A LEITZ MPV 2 PHOTOMETER INCORPORATING AN ORTHOPLAN MICROSCOPE

The light source needs to be stabilized, either by a transformer, a transistorized stabilizer, or by the use of a battery. The lamp should be provided with a heat-absorbing filter. Most lamps have a facility for adjusting the position of the filament.

In addition to the high-power light source for measurement, a low-power source for viewing purposes may be provided.

2. Vertical Illuminator

There are several optical systems which may be employed for reflecting the light beam from the source, so that it passes down the optic axis of the microscope to the objective, and then, after reflection from the object, allows it to pass up the microscope tube to the observation and measuring systems. Of these illuminators, the Berek-type prism which provides three internal reflections is the most widely used for reflectance measurements on coal. The advantages of this reflector include: a far more intense image than is given by glass-plate reflectors; avoidance of the rotation of the vibration direction of light rays that can occur by reflection at a glass plate; and, because three reflections result in a phase difference of 180° , the polarization direction of the incident light is maintained.

However, use of the prism requires that only one half of the aperture of the objective be used for illumination, while the other half transmits the reflected beam. This means that the light will be obliquely rather than directly incident, introducing the possibility of errors in reflectance measurement in polarized light; these can be minimized by setting the polarization direction at 45° to the plane of incidence. Berek (1937) showed that this precaution largely eliminates the variations in measurements on isotropic substances due to the angles of incidence associated with the

aperture of objectives. Broadbent and Shaw (1955) have shown that the same procedure may be justified for readings on uniaxial substances. Several standard methods require or recommend that this setting be adopted when measuring maximum reflectance.

3. Objectives

Objectives of power ranging from X25 to X60, designed for use with polarized light, are suitable for the measurements of reflectance in oil. However, the ASTM standard (1976b) precludes the use of lower power objectives.

4. Stage

Some standard reflectance procedures require that readings be taken on a grid system across the surface of the coal specimen. Where random reflectance measurements are made, this can be achieved with a mechanical stage. Maximum reflectance measurements require a rotating stage to which a small, graduated mechanical stage can be attached.

5. Eyepieces

With X8 or X10 eyepieces the total magnification of the microscope system will be between about 250 and 750. One of the eyepieces may be fitted with a cross hair, micrometer scale, or other reticule to indicate the position and size of the area under measurement. An eyepiece or other magnification system will be located below the photomultiplier. In some apparatus the limiting aperture is placed centrally within this eyepiece at the front focal plane of the eye lens.

6. Monochromatic Filter

Most procedures for the reflectance of coal recommend that measurements be taken in green light with a wavelength of 546 nm. A precision band interference filter may be used for this purpose. It is advantageous to mount

the filter near the photomultiplier tube rather than in a position where it reduces the light to the observer.

7. Limiting Aperture

An aperture is incorporated into the system to limit the size of the field under measurement. Where the aperture has fixed dimensions, it has been possible to place it within the eyepiece below the photomultiplier. Other instruments have variable iris or four-leaf diaphragms located within an alternative magnification system. In some equipment, an image of the illuminator aperture diaphragm is formed on the cathode, so that the area illuminated is not influenced by the size of the limiting aperture. A dispersion screen or negative lens can be used to spread the light over the window of the photomultiplier.

The size of the aperture may be any that is convenient, providing the effective area of measurement lies within the particle and clear of the edges and boundaries with non-vitrinitic materials. The ASTM (1976b) standard specifies an effective spot size of about 5 μm in diameter, and the ICCP (1971) method recommends a field of measurement of 5 to 10 μm in diameter. The author has taken measurements of fields of 20 μm diameter on oriented blocks of vitrain without loss of accuracy. However, if sufficient signal is generated by the photomultiplier, the use of small areas of measurement can reduce the likelihood of an adverse effect from glare or diffraction.

Some optical systems have a facility for observing the measuring aperture and object in coincidence. A swing-in mirror enables the aperture to be illuminated by a small lamp situated on the photomultiplier side of the aperture. Thus, the precise area on which the measurement is taken can be viewed.

Some laboratories have positioned the aperture close to rather than at the optic axis of the microscope. However, although the object stage is seldom perfectly centered, an attempt should be made to restrict the zone of measurement during stage rotation to the central part of the field. Maximum reflectance may be obtained at only two positions during rotation, so that by aligning the system as accurately as possible an operator can be more certain that the field of measurement is represented by a good surface.

8. Levelling Device

Standards and coal blocks or pellets should be mounted with the polished surface parallel to the surface of the stage. This is usually done with a conventional hand press comprised of two parallel plates, the pellet or mount being pressed on to a piece of clay on a glass slide. There are other devices which are particularly suitable for levelling small surface areas (see Piller, 1977); one has three levelling screws which must be adjusted until no variation in focus is experienced during scanning of the surface under the microscope. Screw-wedge tilt adjustment in two perpendicular directions has also been used (McCartney and Ergun, 1969).

The tilt of a surface can be checked by viewing the conoscopic image of the aperture diaphragm during rotation (Galopin and Henry, 1972; Piller, 1977); if it moves, the levelling is imperfect. Jones (1962) has measured the adjustment in focus necessary from one edge of the surface to another, working within a maximum permitted surface slope of 4 minutes.

The parallelism of the plates of the levelling press should be checked and corrected periodically. Even very small misalignments can be seen when the plates are brought close together.

9. Immersion Oil

A nondrying immersion oil of $n_e = 1.518$ is required for reflectance determinations, this value being used in the calculation of the reflectance of the standards (Section IV, F). ASTM (1976b) notes that the temperature at which the oil meets this specification should be 25°C , whereas several of the oils in common use now have a reference temperature of 23°C . Departures in room temperature from 23°C will vary the refractive index of the oil and consequently the reflectance of the material under measurement. Higher temperatures lower the refractive index of the oil, and the reflectance of a standard will be higher than that at the reference temperature. However, providing the standard and coal specimen are close in reflectance, the effect on both of variation in the refractive index of the immersion oil will be similar, and assumption of the 23°C reference value will be valid over the range of ambient temperatures experienced in most laboratories.

Dispersion data for the immersion oils may be available from the manufacturers; temperature coefficients may be obtained from the same sources or measured with a refractometer.

B. Photometric System

Some of the earlier systems for measuring the reflectance of coal employed photocells, but the photomultiplier has become the principal means by which the light intensities are measured. The photomultiplier tubes which are selected should give a high measuring current in the working spectral region and a weak dark current. One photomultiplier tube which has been used extensively is the EMI Type 6094, which has an antimony/casium cathode and eleven dynodes.

A stabilized high-voltage supply is required for the photomultiplier. The manufacturer's or supplier's instructions should be followed regarding the voltage range through which the photomultiplier will provide an acceptable signal/noise ratio. The user of the equipment should satisfy himself that the photomultiplier response is linear through the operating conditions required for the range of reflectance measurements with which he is concerned (see Section IV, E). Response increases with gain to the photomultiplier, and clearly it may be necessary or useful to modify the gain to obtain convenient response to the light from a very small area of a relatively low-reflecting material.

An earthed shield of mu metal placed around the photomultiplier tube may help to prevent interference from magnetic fields. In some climates the provision of air conditioning for the electronic equipment to operate within acceptable limits of temperature may be desirable.

Some photometers are provided with amplification subsequent to the photomultiplier, such as a high-impedance operational amplifier which will enable the signal to the display to be stepped up independently of the gain to the photomultiplier. In this way the noise may be reduced. The amplifier may be provided with a bias control to compensate for the dark current and back reflectance (see Section V, B).

Measurements should be taken in a room with subdued lighting to avoid interference from extraneous sources. The possibility of light entering through the eyepieces in some optical arrangements must be considered.

C. Display and Recording System

This may be a galvanometer, a digital meter, or recorder. Unless used in conjunction with a recording system, the first two require readings to be observed visually and recorded manually. Chart recorders provide a permanent

visual record. Paper tape, magnetic storage, or electronic memory enable the data to be processed following completion of the recording operation.

D. Adjustment of the Microscope System

The manufacturer's instructions concerning the selection of optical components and proper adjustment for reflected-light Köhler illumination should be followed. For ordinary observation, an image of the field diaphragm is stopped down until it is visible within the field of view and par-focal with the object. This can be a useful aid when focusing on glass standards which lack surface detail.

In those systems for which there is no provision for simultaneously viewing the limiting aperture and field of view, there are several ways to ensure that the centers of these coincident. One is to view through the eyepiece and aperture in the phototube before positioning the photomultiplier, locating and noting the coincident point on the object. Another is to move bright spots or dull lines on the object across the central part of the field of view and to note where the photomultiplier gives high or low readings.

The measurement of maximum reflectance involves rotation of the stage, so that errors can be introduced as a result of incorrect levelling and centering, loss of focus, and maladjustment of the stage. Attention to all of these possibilities is required. The amount of variation also can be reduced by adjusting the objective in its centering collar to give a maximum photomultiplier response. However, this adjustment requires that a centering stage be used for the subsequent correction.

In order to avoid excessive vibration to the microscope system, it should be mounted on a sturdy work bench; special provision for further

damping any vibration, like that built into balance benches, can be an advantage.

E. Testing for Linearity of the Photomultiplier

The photomultiplier and recording equipment can be tested for linearity after stabilization of the apparatus (Section V, B). An immersion-oil-covered glass standard should be selected to give a reading near the upper limit of the recorder scale under normal conditions of photomultiplier gain, and at a level of light intensity close to the top of the range to be used. Readings are then taken after reducing the intensity of the light with a range of neutral density filters used singly or in combination. The manufacturer of the density filters will provide transmittance data; the percentage of light transmitted in each case should be proportional to the recorder reading, expressed as a fraction of the original reading (Figure 15). A filter of 50% transmittance should result in a reduction in recorder reading of the same amount. An alternative test is to measure the ratios of intensity values from standards of widely differing reflectances; these should be found to be the same at varied levels of light intensity.

F. Reflectance Standards

Standards which have been used for reflectance determinations on coal include glasses, minerals (e.g., diamond, sphalerite, quartz, zircon, apatite) and synthetic minerals (sapphire, spinel, garnet).

Certain criteria should be satisfied for a standard to be acceptable. These include uniform and isotropic optical properties, chemical stability in water and to the atmosphere, and sufficient hardness and freedom from structural weaknesses and inclusions to take on a uniform and high-quality polish. The reflectance in oil at any desired wavelength should be known

Figure 15. RESULTS OF A TEST OF PHOTOMULTIPLIER LINEARITY

EMI Type 6094B at 1.1 kv; 532 nm. The percentage of light transmitted by a series of filters is proportional to the recorder reading.

or should be calculable from optical data reported by the manufacturer. Standards should be chosen which have reflectances close to those of the coals under examination (Section IV, A, 9).

Where the size of the piece of standard is small, it must be set in an opaque plastic mold. Many laboratories in the United States use sets of up to six glass standards of various reflectances in a single 1-inch diameter mount. Kötter (1960) and others have recommended that the lower face of glass standards be cut at an angle of 30° to eliminate internal reflections. However, petrologists have obtained excellent matchings between standards cut in unmounted cubes which are large enough (~ 1 inch) to be polished by hand. The base and sides of these cubes should be coated with flat black paint to reduce internal reflections. Although multiple standards in a single mount have the advantage of convenience, they tend to develop relief at the boundaries with the plastic--a disadvantage not found with the large cubes.

In the case of glass standards, dispersion curves supplied by the manufacturers can be used to obtain the refractive indices at particular wavelengths. The Fresnel expression (Eq. (7)) is used to derive the reflectances of the glasses in the medium (air or oil) of determined or assumed refractive index.

$$R = \frac{(n_2 - n_1)^2}{(n_2 + n_1)^2} \cdot 100 \quad \dots \dots \dots (7)$$

where R is the reflectance of the standard, expressed as a percentage; n_2 is the refractive index of the glass; and n_1 is that of the medium, taken to be 1 in the case of air, and usually 1.518 in the case of immersion oil.

Piller's (1972) claim, cited by Juckes (1974), that the calculation of the reflectance of glass standards from refractive indices may not be valid because of the influence of surface properties, has led to a suggestion that standards should be calibrated by a qualified laboratory before distribution.

Juckes (1974) has reported that synthetic spinels and garnets have superior polishing and stability characteristics compared to glass. The spinel, yttrium aluminum garnet, and gadolinium gallium garnet which he has used have reflectances in oil of 0.42, 0.92, and 1.73%, respectively.

A particular advantage of synthetic standards is that they do not require repolishing, only a rough rub and clean. Glass standards, on the other hand, require periodic attention. Some laboratories will repolish the standards weekly, even daily; others will leave the standard surfaces under immersion oil until their appearance or matching of reflectance against other standards deteriorates. Special care must be taken during the polishing of standards to avoid even the finest of scratches and also the rapid tarnishing which can result from contact with water (Jones, 1962). The use of distilled water throughout the polishing procedure is recommended. Continued polishing on cloth can produce high relief and orange-peel pitting, which can be minimized by using beeswax laps (Murchison, 1964; Davis, 1971), and corrected by grinding and repolishing.

Because the standards themselves can vary in reflectance owing to the quality of polish or tarnishing, it is essential that the primary standard used be checked against one or more other standards.

Table 1 presents the calculated air and oil reflectance values of several glasses which have been in common use as standards for measurements on coal.

Table I. Optical Properties of
Some Glass Reflectance Standards at 546 nm

Glass	n_e	R_a	R_o ($n_D = 1.515$)
Bausch & Lomb 689309*	1.6945	6.643	0.302
Schott SF 15	1.7045	6.785	0.335
Bausch & Lomb 751278*	1.7572	7.542	0.533
Schott SF 4	1.7617	7.607	0.552
Schott SF 11	1.7919	8.045	0.685
Chance VB 15/137	1.8095	8.302	0.767
Schott SF 6	1.8127	8.348	0.783
Chance VB 140/62	1.8152	8.385	0.795
Bausch & Lomb 850324*	1.8564	8.989	1.006
Bausch & Lomb 915213*+	1.9255	10.008	1.400
Chance F 2/14	1.9290	10.060	1.422
Bausch & Lomb 980222*	1.9908	10.974	1.815

* In the United States, combinations of these standards have been embedded in single mounts.

*+This standard may give a lower reflectance than the calculated value which appears in this table.

V. PROCEDURES FOR THE DETERMINATION OF REFLECTANCE

A. Sample Preparation

The principal objective of the sample preparation procedure is to prepare for microscopic examination a suitable polished surface which is representative of the coal under consideration. This usually involves crushing the coal, embedding it in a cold-mounting medium, and grinding and polishing a surface, which, as specified by ASTM (1976a), should meet the following requirements:

- (i) The entire surface should be flat, and have little or no relief between the various coal components or at the edges of coal particles.
- (ii) It should be free of pits caused by loss of coal grains.
- (iii) It should appear substantially free of scratches under the microscope.
- (iv) It should be clean and free of grinding and polishing compounds.

Providing the above requirements are satisfied, there is a wide latitude of embedding and polishing equipment, materials and procedures which can be employed. A common embedding procedure in the case of particulate coal samples is to mold about 8 g of about minus 700 μm [ASTM, (1976a) specifies minus 850 μm (20 mesh)] into a 1-in.-diameter pellet. It is convenient to use just sufficient binder to hold the particles together in order to reduce the time spent moving between coal particles in the course of microscopic study and to reduce gravity segregation.

Different laboratories have used aluminas, diamond paste, and chrome, cerium, and magnesium oxides as polishing compounds. Silks, and low-nap wool, cotton, and synthetic cloths have all been used as lap coverings.

Polishing can be done by hand on stationary or rotating laps, or with automatic polishing devices. Excessive polishing produces relief and should be avoided.

Harrison (1965, 1966) reported that the reflectance values of some coals can be significantly affected by moisture content. Consequently, pellets should be kept in a desiccator for some hours [ASTM (1976b) specifies 15 h] prior to measurement. However, because low-rank coals tend to slack under dry conditions, careful compromise is needed for these coals.

B. Reflectance Measurement

The microphotometer should be set up, adjusted and tested as described in Sections IV, D and E. Other checks, adjustments and standardization procedures are required on a day-to-day basis as part of the measurement protocol.

A warm-up period of about 15 min to 1 h is needed for the equipment. For convenience in routine operation, the equipment may be activated by a time switch.

The scale for reflectance measurement is set with two end values, one being zero when no light impinges on the photomultiplier, and the other being a value to represent the reflectance of the primary standard for a set of determinations. Zero is set using the zero control of the amplifier or display unit. The photomultiplier is then exposed to light from the surface of the carefully leveled standard, and the known reflectance value for that standard (or some simple function of it) is set on the display or recorder scale by adjusting the range control of the display unit and the photomultiplier or amplifier gain controls. During a full rotation of the stage, the standard reading should not vary by more than about $\pm 0.01\%$ reflectance.

A small amount of the incident light is reflected from the back lens of the objective. In reflectance measurements, this glare (back reflectance) can be significant, so that a simple correction may have to be applied. Back reflectance may be measured using a polished pellet of opaque resin or powdered coal (see Jones, 1962) in which a vertical hole about 3 mm in diameter and 5 mm deep is drilled. After filling the hole with immersion oil and placing the pellet on the microscope stage, the polished surface around the hole is focused while the photomultiplier shutter remains open. The pellet is then moved so that a reading is taken directly over the hole; the value obtained is the back reflectance. If this value is significant, the zero control of the amplifier or display unit can be set back to zero while the photomultiplier shutter remains open. Alternatively, the original standard may be replaced and the gain of the photomultiplier or amplifier adjusted so that the standard reflectance reading is increased by an amount equal to the back reflectance; this same value then should be subtracted from all subsequent readings or their average. Some workers have used an inclined oil-covered glass cover slip to measure back reflectance (Kötter, 1960); others have employed a "black glass" standard to obtain a zero reading.

The choice of primary standard should be determined by the reflectance level of the coal, which should be checked after the initial standardization. Juckes (1973) has observed that errors due to variation in the refractive index of the immersion oil with temperature are least when a standard is used which has a slightly higher reflectance than that of the coal.

Once a direct-reading scale has been established with a correct zero and a primary glass standard, the reflectance of one or more secondary standards should be determined as a check. It is common practice for

petrographers to work within a tolerance of $\pm 0.02\%$ absolute of actual standard values. Other petrographers (see Jones, 1962; Davis, 1971; Juckes, 1973) have used a relative-percentage tolerance of $\pm 1\%$ of the standard value. For each standard, the reflectance should be checked at two or more positions, all near the center of the surface.

After standardization, reflectance readings are taken on the vitrinite in the coal pellet (particulate block). The types of vitrinite which may be selected for measurement were discussed in Section I, A. For most applications it is desirable to select particles according to a grid system; the x and y scales provided on the mechanical stage attachment should be used so that the pellet surface is effectively covered in the course of 50 to 100 readings. Readings are taken when the reference point in the eyepiece reticule coincides with a particle of vitrinite. Adjustment of the stage is often necessary so that the reading is taken on a defect- and relief-free area away from the boundary of the coal particle. The focus is corrected and the photomultiplier shutter opened. For random reflectance measurements, the reading is taken without rotation; for maximum reflectance measurement, the stage is steadily rotated through 360° . The speed at which the stage is rotated depends upon the response time of the recorder or meter. According to ASTM (1976b), the rotation rate should be about 4 rpm. Care must be taken to avoid using too much hand pressure on the stage during rotation because this could result in loss of focus or centering. Usually, the highest value read during rotation is taken as that representing maximum reflectance, although some petrographers average the two maximum values occurring at 180° to each other.

In the course of a series of measurements on a coal sample it may be necessary to check the zero and standard reading. Depending upon the stability of the equipment at the time of operation, such a check might be required after as few as five or as many as 50 readings. In any event, the calibration should be checked every 20 to 30 min, and at the completion of readings on each pellet. If the reading on the standard is not within the accepted limits of tolerance, then the preceding group of readings is rejected. A few petrographers might correct an accepted group of readings for any drift in the standard reading which has occurred. Following the calibration check, any readjustment to the precise standard reflectance value should be made.

One hundred measurements of reflectance are thought to be necessary for a single coal-seam sample; often these are composed of sets of fifty readings on each of two pellets. According to ICCP (1971), no significant increase in precision is gained by moderate increase in the number of readings. In order to obtain a good distribution of vitrinite reflectance from a blend of coals, the ICCP recommends that 500 readings be taken, 250 from each of two pellets.

C. Calculation and Reporting of Results

Reflectance is usually reported as the mean of the individual readings of maximum or random reflectance. Where more than one pellet has been read for a sample, the means of approximately equal numbers of readings on each pellet may be averaged. The standard deviation of the measurements is often reported.

Often, the petrographer or coal user will be interested in the reflectance distribution of a coal or coal blend. This may be reported as a table or histogram showing the numbers or percentages of readings at various

reflectance levels. In North America, considerable use is made of the distribution of so-called Vtypes. Each Vtype represents a reflectance range of 0.1%; for example, all readings in an analysis between 1.50 and 1.59% are assigned to Vtype 15. Figure 16 is an example of a Vtype distribution in the form of a computer-generated histogram.

Another common form of histogram is that which groups reflectances into 1/2-Vtype classes (Figure 17). A range of reflectance from 1.50 to 1.54% represents one such class, designated by the mid-point expression 15.25. The reporting of results in this manner is particularly useful in demonstrating the contribution of coals of different ranks in a blend (ICCP, 1971).

Reproducibility of the mean should be $\pm 0.02\%$ reflectance at the 95% confidence level for representative samples of a coal.

VI. INNOVATIONS IN REFLECTANCE DETERMINATION

Manual rotation of the microscope stage is, at best, unsteady; at worst, clumsy and a source of error. Methods of motorizing this operation have been developed (e.g., Melvin, 1973), but have not been widely adopted. Melvin's system was designed to provide an even speed of operation and a rapid start-stop capability through an electromagnetic clutch. Lateral movement of the microscope stage was minimized through the use of a toothed nylon-belt drive.

There is now available a variety of data processors which will store and process the readings obtained during reflectance analysis (see for e.g., Jacob *et al.*, 1970). Where maximum reflectance is read, there is a particular advantage in using a digital voltmeter equipped with a peak detector (Spackman *et al.*, 1973). This unit holds the maximum value obtained during each stage rotation. Operation of a switch can transfer the reading to tape

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REFLECTANCE ANALYSIS FOR PSOC-496

TOTAL COUNTS ON VITRINITE + PVIT = 130
MEAN-MAX RO = 0.582
PELLET 1 = 0.579
PELLET 2 = 0.586
STANDARD DEVIATION = 0.037

OPERATOR: ARV
STANDARD = 0.533, 1.006

FREQUENCY TABLE FOR VITRINITE + PVIT

	LEVEL OF RO	% OF TOTAL	REFLECTANCE HISTOGRAM:
1	0.49	1.00	0
2	0.50	2.00	00
0	0.51	0.00	
3	0.52	3.00	000
5	0.53	5.00	00000
6	0.54	6.00	000000
4	0.55	4.00	0000
10	0.56	10.00	0000000000
7	0.57	7.00	0000000
16	0.58	16.00	0000000000000000
11	0.59	11.00	00000000000
11	0.60	11.00	00000000000
6	0.61	6.00	000000
5	0.62	5.00	00000
3	0.63	3.00	000
5	0.64	5.00	00000
3	0.65	3.00	000
0	0.66	0.00	
1	0.67	1.00	0
1	0.68	1.00	0

VTYPE TABLE FOR VITRINITE + PVIT

VTYPE	COUNTS	% OF TOTAL
4	1	1.00
5	64	64.00
6	35	35.00

Figure 16. COMPUTER PRINTOUT OF REFLECTANCE DATA AS A HISTOGRAM AND
VTYPE DISTRIBUTION

Figure 17. REFLECTANCE HISTOGRAM OF A BLEND OF HIGH-VOLATILE COALS

for subsequent processing. The unit is then cleared for the next reading; scanning between measuring points is done while the photomultiplier shutter is closed.

Bennett (1969) has described a system in which the incident-light polarizer is rotated instead of the stage; this is possible because introduction of a plane-glass elliptical compensator has corrected the adverse effect of partial plane polarization of the light beam due to the source. Melvin (1973) was unable to overcome this problem satisfactorily with a compensator; his approach was to mount a co-rotating lamp and polar vertically above the objective. A portion of the reflected light was directed to the photomultiplier located at the normal position for the light source. Ting (in press) has described a three-azimuth method of determining the maximum reflectance of coal.

The more intense light of laser sources may aid microphotometry by enabling stable signals to be obtained from very small areas. BCRA (1976) has worked with a helium/neon laser to obtain reflectance measurements on areas as small as $0.5 \mu\text{m}$. A modification to the accepted technique for coal may be required because of the wavelength involved (632.8 nm). Problems with ghost reflections and interference effects may have to be overcome (Kaye, 1976).

A new development in reflectance measurement may result from research into radiometric systems. David and Galbraith (1975) have described the design of the Denver Universal Microspectroradiometer (DUM) which uses direct photoelectron counting for precise optical measurements on biological materials. As noted by these authors, advantages of photon counting include the precision resulting from a digitized signal, and minimization of errors attributable to the analog components of the system. The U.S. Steel laboratories

are currently investigating photon counting for conventional and automated reflectance measurements on coal, and early results are encouraging (Gray, 1977).

As a rapid means of checking the calibration of reflectance equipment, Bennett (1969) has used a bypass tube to utilize that portion of light normally cut out by the prism of the incident illuminator. Once the light intensity of this direct comparison beam has been matched against that reflected from the standard by rotating a disc of polaroid, it can be checked at any time without reintroduction of the standard.

VII. AUTOMATED REFLECTANCE MICROSCOPY OF COAL

Petrographic analyses are time consuming and often highly subjective. Though subject to its own inherent problems, automated reflectance microscopy should be rapid and objective. Ideally, it could be used to obtain quantitative characterization of coal so that routine quality-control analysis can be handled quickly and without the need for trained and patient technicians.

Probably the earliest attempt at an automated method of reflectance measurement for coal was that of Vendl (1934). He described how the stage of a microscope was driven by clockwork; the same motor turned a cylinder covered with light-sensitive paper to record the deflections of a mirror galvanometer. The volume percentages of mineral components were estimated, and Vendl predicted that the approach might be applicable to the determination of the organic components.

Coal petrographers applied the AMEDA (Automatic Microscope Electronic Data Accumulator) system to measure pyrite size distributions, the proportions of arbitrary reflectance categories of constituents, and coke pore volumes (Bayer *et al.*, 1968) following development of a prototype which

has been described by Bomberger and Deul (1964). Size categories were assigned to 10 electronic counters. The equipment was used to measure the relative amounts of reactive and inert petrographic entities in coal.

Workers at the U.S. Bureau of Mines (McCartney and Ergun, 1969; McCartney *et al.*, 1971) recorded data from their automated system on magnetic tape for computer processing. A means of scanning a spiral path was developed to provide a smooth motion at constant velocity, in order to overcome the jerky movement and loss of focus which can attend linear traverses. The circular fields of successive adjacent measurements were 2 μm in diameter. The speed of traverse employed in the course of their reported experiments was 177 $\mu\text{m}/\text{sec}$; an analysis involving 1 million readings took about 3-1/2 h, but they predicted that the speed of operation could be increased by as much as tenfold. Pyrite contents could be determined nearly as well as by visual microscopy, but the size distribution of pyrite was considerably affected by discontinuities on the pyrite surfaces, resulting in reduced apparent size. Use of the same equipment for performing automated petrographic analyses gave fairly good agreement with the results of visual microscopy, especially in demonstrating the difference in exinite and vitrinite contents of dull and bright coal. Eight reflectance ranges for the coal components were determined by prior examination, but subsequently arbitrary adjustment of these values was found to be necessary.

At The Pennsylvania State University, an automated reflectance microscope (ARM or Rapid Scan) system has been developed which incorporates some of the ideas originated in the AMEDA and U.S. Bureau of Mines systems (Davis and Vastola, 1977). The equipment (Figure 18) is based on a Leitz Ortholux microscope--MPV 1 photometer interfaced with a Digital Equipment PDP-9 computer. Stage movement is provided by a modified Femco AMEDA drive

Figure 18. SCHEMATIC DIAGRAM OF THE AUTOMATED REFLECTANCE MICROSCOPY (ARM) SYSTEM USED AT THE PENNSYLVANIA STATE UNIVERSITY

unit controlled by the computer. The stage speed and data sampling time are selected so that a recording is made for every scan distance of 2 μm . Since the effective area of measurement determined by the limiting aperture is 2 μm x 2 μm , a series of readings is taken on adjacent fields. A computer array of 1,023 elements is used to accumulate the data.

The Pennsylvania State University's ARM system matched the ability of visual analysts in distinguishing between relatively coarse and fine pyrite distributions in runs involving approximately 400,000 individual readings on each pellet, an operation requiring about 18 min. Repeat runs on individual pellets showed reasonable reproducibility of pyrite chord-size distributions, although the sampling of a few exceptionally large particles could affect the particle size at 90% undersize. The amount of pyrite expressed as a percentage of linear scan was very reproducible. Davis (1975) showed that pyritic sulfur contents obtained by this method gave results which are reasonably consistent with values determined by wet chemical analysis.

Problems in the use of the ARM system for pyrite size distribution were outlined by Davis and Vastola (1977). They include the small particle size of coal which presently can be analyzed, and the statistically small number of pyrite particles which are encountered. For example, in the course of a 50-cm scan, there might be 300 to 600 particles of pyrite traversed. The system also cannot resolve particles in the smallest size range, namely below 2 μm .

Vitrinite-rich coals give well-defined ARM peaks in the vitrinite reflectance range when non-polarized light is used. The reflectograms of a series of coals of varied rank (Figure 19) enables the modal values to be read precisely. If the assumption is made that these peaks represent a

Figure 19. REFLECTOGRAMS OF A RANK SERIES OF HIGH-VITRINITE COALS OBTAINED WITH AN AUTOMATED REFLECTANCE MICROSCOPE
The lower reflectance peaks represent readings on pellet binder.
(from Davis and Vastola, 1977)

normal distribution of vitrinite reflectance, then the modal values should be close to the mean random reflectances of the samples. A factor to allow for the anisotropy at any rank level can be added in order to calculate maximum reflectance (Hoover *et al.*, 1976; Davis and Vastola, 1977) as shown in Table II. The results obtained are compared with values determined by the conventional reflectance technique. Although the differences shown for these provisional automated analyses might not be acceptable for some purposes, they encourage further consideration of this approach as a rapid means of measuring coal rank.

The profiles of ARM reflectograms provide some assessment of petrographic composition (Davis and Vastola, 1977). Figure 20 shows the reflectograms of three lithotypes selected from one high volatile bituminous seam. The maceral-group compositions, determined by visual petrographic analysis, are shown as bar graphs arranged in order of increasing reflectance to the right, matching the order in which the maceral groups would be represented in a reflectogram. For these three samples, the heights of the vitrinite peaks in the reflectograms correspond to the values for this maceral in the bar graphs. The frequency of readings in the higher and lower reflectance sections of the reflectograms likewise correspond with the concentrations of exinite and inertinite, respectively. The proportions of the three maceral groups in lithotype PSOC-2D are approximately the same, and this is indicated by the relatively flat profile of its reflectogram.

There has to be a balance between effective sampling of the signal and smearing of results. The Pennsylvania State University's ARM uses a very small sampling time, and conditions the analog output by a low-pass filter to improve the signal/noise ratio. In order to gain the extra resolution

Table II. Comparison of Maximum Reflectances Derived from
Automated and Conventional Techniques
(after Davis and Vastola, 1977)

Sample no.	Random Oil reflectance (%) (automated, unpol.)	Max. oil reflectance (%) (automated, calc.)	Mean max. oil reflectance (%) (polarized)
PSOC			
242	0.40	0.40	0.43
22	0.48	0.48	0.51
233	0.55	0.55	0.61
125	1.19	1.19	1.17
320	1.37	1.37	1.30
236	1.40	1.40	1.45
127	1.53	1.57	1.53
318	1.57	1.65	1.65
85	2.50	2.77	2.82
159	2.81	3.19	3.10

Figure 20. COMPARISON OF AUTOMATED REFLECTANCE MICROSCOPE REFLECTOGRAMS WITH PETROGRAPHIC ANALYSIS

The vertical scale is based on the most frequent reading, with a tenfold factor for reflectances less than 0.12%.

Ex., exinite; Vit., vitrinite; In., inertinite

The lower reflectance peaks represent readings on pellet binder.

(from Davis and Vastola, 1977)

required for effective quantification of petrographic composition by automated means, further work is being undertaken using a pulsed scanning stage.

Petrographers at Nippon Steel Corporation (Kojima *et al.*, 1974; Nippon Steel Corporation, 1976) have developed an automated reflectance microscope with a pulsed motor which traverses in steps to 10 μm . They interpret the reflectogram (Figure 21) by fitting a normal distribution for vitrinite reflectance. An area is assigned to represent readings on the pellet binder. Contributions made by other maceral groups then are apparent. The reflectograms obtained by these workers contain a number of well-defined peaks, which they have attributed to inertinite. However, the discontinuous and regularly spaced form of these peaks raises some doubt about this interpretation.

No attempt has been made in this chapter to discuss developments in automated microscopy in areas other than the petrography of coal. Nor will a review be made of the study of coal by image-analysis systems which do not employ the measurement of reflectance as a means of discrimination, although such methods have been employed to measure the size distribution of pyrite in coal (Harris *et al.*, 1977; Lebedzik and Dutcher, 1977), and for quantifying the texture of coke (BCRA, 1975). Harris (1977) has established a correlation between reflectance and the threshold control setting of an image analyser, enabling him to interpret the results of field analyses as reflectograms.

Shibaoka (1974) has suggested using vacuum-coating techniques to impart different colors to the various coal macerals. The differentiation which would result by measuring reflectance at two different light wavelengths could be usefully employed in automated methods of maceral analysis.

Figure 21. REFLECTOGRAM OBTAINED WITH THE AUTOMATED REFLECTANCE MICROSCOPE SYSTEM USED AT NIPPON STEEL CORPORATION

B, binder; E, exinite; V, vitrinite; SI, semi-inertinite; I, inertinite
(from Kojima *et al.*, 1974)

VIII. APPLICATIONS OF REFLECTANCE DETERMINATIONS

The basic use to which determination of vitrinite reflectance is applied is the measurement of the degree of metamorphism or rank of coals and the organic components of sediments. The advantages of this determination are: i) that it is rapid, and ii) that unlike most chemical indices used for this purpose, it is free of the influence of petrographic variability. McCartney and Teichmüller (1972) have observed that vitrinite reflectance is the best single parameter of rank for coals with less than 30% volatile matter, and it is nearly comparable to other parameters for high volatile bituminous coals.

Using established curves, it is possible to relate the reflectance of a coal to parameters such as carbon content, volatile matter (Figure 22), and calorific value. However, consideration needs to be given to the methods used in the determinations. For example, volatile matter is determined at different temperatures in different national standard methods, and reflectance values may be the means of maximum or random measurements.

McCartney and Teichmüller (1972, 1974) have suggested the random reflectance limits in Table III as guides to ASTM rank. Based on measurements on 140 coals at The Pennsylvania State University, the limits of ASTM rank categories appear to coincide approximately with the maximum reflectance values given in the same table. The ranges of three important classes of United States coals as identified by maximum reflectance at Bethlehem Steel Corporation also are included.

Two-parameter classification systems have been developed which use reflectance as a parameter of rank. Bennett and Taylor (1970) have proposed a scheme in which the second parameter, for the petrographic composition, is the sum of vitrinite-rich microlithotypes. Subsequently, Strauss *et al.* (1976) used a similar approach, but with reactive-maceral content (vitrinite

Figure 22. REFLECTANCE-VOLATILE MATTER RELATIONS FOR EUROPEAN AND UNITED STATES COALS
(from McCartney and Teichmüller, 1972)

Table III. Oil Reflectance Limits of ASIM Coal Rank Classes

Rank	Maximum Reflectance, %	Maximum Reflectance, %†	Random Reflectance, %*
Subbituminous	-0.47		
	C 0.47-0.57		
High Volatile Bituminous	B 0.57-0.71	<1.03	0.50-1.12
	A 0.71-1.10		
Medium Volatile Bituminous	1.10-1.50	1.03-(1.35-1.40)	1.12-1.51
Low Volatile Bituminous	1.50-2.05	> (1.35-1.40)	1.51-1.92
Semianthracite	2.05-3.00 (approx.)		1.92-2.50
Anthracite	>3.00 (approx.)		>2.50

† Procedure of Bethlehem Steel Corporation using reactive vitrinite reflectance.

* After McCartney and Teichmüller (1972, 1973).

plus exinite) as the petrographic index, to relate these basic properties to others such as swelling index, calorific value, and volatile matter (Figure 23). Davis *et al.* (1976) related these same parameters to the liquefaction potential of coals. Eremin and Kosina (1976) have proposed a classification using the percentage of inertinite as an index of maceral composition. Marshall (1976) has developed a classification system which uses reflectance and dilatation as measured in the Ruhr dilatometer to assign 4-digit classes (Figure 24). A similar approach has been followed by Okuyama *et al.* (1970), using reflectance and Gieseler fluidity. This is discussed below in connection with predictions of coking behavior.

Several laboratories use petrographic analyses to predict the properties of coke which would be produced from a subject coal. The reasoning is the same as that on which the classification systems just discussed are based--that rank and petrographic composition are independent factors which together determine a coal's behavior. Only coals within a certain range of bituminous rank display the kinds of plastic behavior which is desirable for metallurgical coke manufacture; within this range the coking properties vary, with a maximum development at about 88-89% carbon. The properties of coke formed from coals of any one rank, given uniform coking conditions, depend upon the petrographic composition, because only certain macerals become plastic when the coal is heated.

Schapiro *et al.* (1961), following the approach of Ammosov *et al.*, (1957) use data similar to those displayed in Figure 25 to calculate the optimum inert contents for a coal blend. They assume that all of the different Vtypes or reflectance levels read in the course of an analysis, even in a single coal, behave in a different fashion, and require different optimum levels of inert macerals to give the maximum coke strength for that Vtype. Weighting

Figure 23. RELATIONSHIP BETWEEN COAL PETROGRAPHIC INDICES AND CALORIFIC VALUE (SOLID LINE) AND VOLATILE MATTER (BROKEN LINE)
(Modified from Strauss *et al.*, 1976)

Figure 24. A PROPOSED COKING COAL CLASSIFICATION SYSTEM
 (from Marshall, 1976)

Figure 25. RELATIONSHIP OF STRENGTH INDEX, VTYPES AND INERT CONTENT OF COALS
(from Schapiro and Gray, 1964)

these optimum inerts according to the proportions of the corresponding Vtypes present enables the optimum inert content for the whole sample to be calculated. The ratio of the actual inert content of the coal to the optimum inerts is referred to as the balance index. A second index is derived using the data displayed graphically in Figure 25; from the actual inert content of the sample, the strength index is calculated according to the proportions of the different Vtypes present. The two indices are used to obtain a predicted ASTM coke stability from a set of curves like those in Figure 26.

Smith (1973) uses the proportions of vitrinite reflectance classes to calculate Micum 40 indices of cokes from coals or blends used by the National Coal Board. The other principal compositional parameter is the total amount of inert constituents (inertinite + "ash"). Corrections are applied which allow for the beneficial effect of greater amounts of inert constituents in the finest (minus 0.12 mm) coal size fraction, and the detrimental effect of increased inerts in the coarsest (plus 3 mm) fraction.

The method of coke-strength prediction used in the laboratories of Bethlehem Steel Corporation (Benedict *et al.*, 1968) recognizes only two vitrinite types in any coal--"reactive vitrinite" and "pseudovitrinite." Sets of curves such as those in Figure 27 relate the inert content of the sample, the mean maximum reflectance of the "reactive vitrinite", and ASTM coke stability. Other curves exist for determining the stability of cokes from coals of other ranks and blends of high and low volatile coals.

The pseudovitrinite in a coking coal is considered to act as an "effective inert." Therefore, a proportion of the pseudovitrinite is added to other inert macerals (arbitrarily taken as micrinite + macrinite + fusinite + 2/3 semifusinite + mineral (vol. %, calculated)). Curves such

Figure 26. BALANCE AND STRENGTH INDICES AND EMPIRICALLY-DETERMINED ISOSTABILITY LINES
(from Schapiro and Gray, 1964)

Figure 27. BETHLEHEM STEEL CORPORATION CORRELATION CURVES BETWEEN PETROGRAPHIC COMPOSITION AND ASTM STABILITY FOR PREDICTING THE STABILITY OF COKE PRODUCED FROM HIGH- AND MEDIUM-VOLATILE COALS
(from Benedict et al., 1968)

as those in Figure 28 are used to determine the proportion of the pseudo-vitrinite which should be considered as an effective inert; this proportion increases as the mean maximum reflectance of the pseudovitrinite increases above that of the reactive vitrinite.

The predictive methods described above are meant to apply strictly to the coking operations associated with the laboratories which developed them, although modifications to allow for other carbonizing conditions have been investigated. Mackowsky and Simonis (1969) have developed equations for the calculation of the Micum 40 and 10 indices involving such coking parameters as coal size consist, oven width, bulk density, coking rate, and coking time. This system requires conversion of maceral and reflectance data to a dilatation index and volatile matter as determined in the German standard method.

Okuyama *et al.* (1970) have used reflectance and Gieseler fluidity to investigate the blending of coals utilized in the coking operation of Nippon Kokan K.K. (Figure 29). They have identified their acceptable coking coal blends as having a mean reflectance of about 1.2% and a Gieseler fluidity of about 60 to 1000 dial divs/min. As many as 20 coals of varying ranks and fluidities may be combined in these Japanese blends. Lower mean reflectances of the blends have been shown to result in physically weak coke; too high a fluidity renders the coke liable to cracking--if too low, the adhesion between particles will be poor. Blend components from quadrants I and IV of Figure 29 are important in maintaining the mean rank of the blend above the lower limit of mean reflectance, and components from quadrants I and II maintain the mean fluidity.

In addition to predicting coke-strength characteristics from reflectance and other data, other important carbonization properties have been

Figure 28. BETHLEHEM STEEL CORPORATION CHART FOR DETERMINING THE DEGREE OF INERTNESS OF PSEUDOVITRINITE IN HIGH-VOLATILE COALS
(from Benedict et al., 1968)

Figure 29. REFLECTANCE AND FLUIDITY OF COALS USED IN JAPANESE COKING BLENDS
(from Okuyama *et al.*, 1970)

related to reflectance. Pressures above 2 psi are reported to damage coke oven walls, and Bethlehem Steel workers (Benedict and Thompson, 1976) have shown how the peak pressure exerted varies with the reflectance of "reactive vitrinite". Figure 30 illustrates this relationship for low volatile bituminous coals, and shows how increased inert contents can lower the coking pressure. Graphical data also have been published which relate reflectance to coke reactivity (Thompson and Benedict, 1975; Okuyama *et al.*, 1970).

Some steel companies map their coal properties using iso-reflectance lines in addition to other relevant properties. The maps provide information on the quality of coke that would be produced from coals of different subdivisions within a mine, and aid in the development of plans for mining coals consistent with the company's coke-oven blending practice (Thompson and Benedict, 1976).

Reflectance measurements on the dispersed organic matter in borehole and other sections of sedimentary rocks is providing a means of studying past geothermal gradients, coal metamorphism, and the likelihood and nature of associated petroleum occurrences (see Hood *et al.*, 1975). Table IV, after Bostick and Damberger (1971), is a composite of correlations between rank parameters, including reflectance, and the occurrence of hydrocarbons in associated strata.

Other uses to which reflectance measurements have been put include the estimation of the chemical properties of fresh coals from the reflectance of weathered specimens (Figure 31) (Chandra, 1962), and the estimation of temperatures to which coals have been exposed in natural and industrial processes. Calculations of physical-constitution parameters have been reported by Schuyer and van Krevelen (1954), Leicester (1956), Bond *et al.* (1958) and Davis (in press), although the theoretical bases of these calculations are

controversial. The parameters include the molar refraction which has been used to derive the number of aromatic carbon atoms, total carbon atoms, and number of rings per mean structural unit of vitrinite.

Table IV. Coalification Stages Based on Various Parameters of Rank (Left)
Correlated with the Occurrences of Hydrocarbons in Associated Strata*

Refl. % R _o	Fixed carbon %	ASTM coal classes and groups	Eastern United States	Canadian Great Plains	North Texas, Paleozoic	Oklahoma, Carbonif- erous (Relative chance of finding oil)	Eastern U.S. and Midcontinent, Pennsylvanian	Kentucky, Carbonif- erous	Northwest Germany, Mesozoic, Paleozoic	Review of world picture
0.3		Lignite		1 Wet						
0.4	50	C Subbituminous	Commercial	or dry	Principal oil fields	100	Mixed base below 35*	Nearly all oil-gas production	Oil fields in lower Saxony	Methane formation in early catagenesis
0.5	55	B Subbituminous		2 hydro-						
0.6		A Subbituminous	oil		Principal fields of light oil and gas	10	Mixed base 35-40*			Oil formation (plus gas)
0.7	60	C High-volatile	fields	carbons	Rare high- gravity oil; common gas fields	1	Paraffin base above 40*	Most gas pools	Gas only	
0.8	65	B High-volatile		3						
0.9		A High-volatile			Usually no commercial production					
1.0	70	Medium-volatile	Oil deadline							
		Medium-volatile		Dry	No oil or gas, with rare exceptions		Dry gas only			Wet gas and gas condensates
	80	Low-volatile	No commercial oil fields; gas fields may occur	4 gas						
2.0		Semi-anthracite		Dry gas or barren			No hydro- carbons			Late catagenetic methane
	90									
3.0		Anthracite								

*Modified from a compilation of Bostick and Damberger (1971) of the studies of other workers in the geographical areas cited.

Figure 30. CURVES USED TO IDENTIFY PRESSURE CHARACTERISTICS OF LOW-VOLATILE COKING COALS
(from Benedict and Thompson, 1976)

Figure 31. RELATIONSHIP BETWEEN REFLECTANCE AND CARBON CONTENTS OF NORMAL AND WEATHERED COALS
(from Chandra, 1962)

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