CARBONIZATION MEASUREMENTS OF POLLEN-GRAINS AND SPORES AND THEIR APPLICATION

BY

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SUMMARY

This study presents the equipment and technique used in the author's palynological carbonization studies. The equipment consists mainly of modified Zeiss microphotographic equipment. A photoelectric cell fitted with a field restrictor is used to measure the light absorption of a spore or pollen type. The light-absorption value is considered to be a measure of carbonization, and values of a standard spore or pollen type are used to construct a carbonization line for an individual well.
Isocarbonization lines can be constructed in sections to show local or regional carbonization. Maps, block diagrams, panel diagrams, etc., can be constructed with either isocarbonization or carbonization contours.
Carbonization is a low-grade metamorphic process; an understanding of this process and its effects can be useful to the petroleum industry. The primary advantage of the described technique is that the degree of carbonization of pollen-grains and spores is a measure of low-grade (organic) metamorphism in areas or intervals devoid of coal. In addition the technique is cheaper and faster than the usual chemical coal analyses. Samples normally taken in the petroleum industry, such as cores, sidewall cores, and cuttings, can be used in palynological carbonization studies.
Palynological carbonization studies may be undertaken not only in strata in which palynological time correlations are possible but also in areas or sections in which alteration is too great for routine palynological time correlation. The carbonization method is unsuccessful in extremely carbonized zones, where alteration makes standard types unrecognizable, or, of course, in strata devoid of pollen-grains and spores.
The equipment and technique described here yield reasonably accurate and reproducible data that may shed some light on the temperature history of the organic material, the sediments, and the associated hydrocarbons.

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INTRODUCTION

Pollen-grains and spores in coals from different ranks, studied under the microscope (transmitted light) after maceration methods or in thin sections, gradually change in color from yellow to light brown to dark brown-black. Changes in color of pollen-grains and spores from other sedimentary rocks, such as shales, sandstones, etc., are similar. The term carbonization is used in this study to denote this alteration process.

The chief interest and importance of carbonization investigations of pollen-grains and spores is that the carbonization measurements of pollen-grains and spores are indices of the level of organic metamorphism. In order to avoid confusion with the definition of metamorphism accepted by hard-rock geologists, the term organic metamorphism is suggested in palynological carbonization studies.

The main objective of this investigation is the quantitative determination of the level of organic metamorphism in sediments. The knowledge of the level of organic metamorphism in sediments is important for several reasons. In the first place, a reasonable to optimum preservation of pollen-grains and spores is a necessity for palynological correlation and/or ecological studies. Strongly to intensely carbonized pollen-grains and spores are opaque and show no structure. They are, therefore, difficult to recognize and palynological correlations and/or ecological investigations in areas or intervals in which they occur are difficult and often impossible. Because severe carbonization is a limiting factor in carrying out palynological or ecological studies, it is obvious that the delineation of severely carbonized areas or intervals is important to the palynologist and to the exploration effort in general.

In the second place, the carbonization degree of pollen-grains and spores is an index of the level of organic metamorphism to which also other organic matter (other than pollen-grains and spores) in the sediment has been subjected including oil. It is probable that important economical oil accumulations will diminish progressively as the intensity of organic metamorphism alteration increases.

Another reason why the determination of the level of organic metamorphism may be of importance is the possible correlation between sediment properties and the carbonization of pollen-grains and spores.

In order to determine the level of organic metamorphism, two methods have been developed. The simplest and fastest of the two methods to measure the degree of carbonization of pollen-grains and spores is the subjective evaluation of the color of the pollen-grains and spores by their appearance under the microscope (transmitted light). Several attempts were made to standardize the visual color determination. Reference slides of pollen-grains and spores of different ranks were compared with pollen-grains and spores to be measured. Color charts and color sticks were devised as standards of carbonization values. These

attempts were unsatisfactory for detailed investigations because the inherent disadvantage of using color standards. The difficulty is the visual comparison of subtle color differences.

Color changes of pollen-grains and spores with increasing low-grade metamorphism are accompanied by a decrease of translucency to final opacity as observed with transmitted light. As a result of this observation, photoelectric measurements of the translucency of pollen-grains and spores have been made in order to express the carbonization degree of pollen-grains and spores objectively.

Terminology

Most of the following definitions of technical terms, used in this study, have been compiled from *Glossary of Geology and Related Sciences* (American Geological Institute, 1960), *Pollenmorfologiske definitioner og typer* Inversen and Troels-Smith, 1950) and the *International Handbook of Coal Petrography* (International Committee for Coal Petrography 1963).

British thermal unit: Abbreviated B.t.u. A unit of heat which is 1/180th part of that required to raise the temperature of one pound of water from 32 °F to 212 °F at sea level. Usually considered as that amount of heat required to raise the temperature of one pound of water from 63 °F to 64 °F.

Carbon ratio: The ration of the fixed carbon in any coal to the fixed carbon plus the volatile hydrocarbons; expressed in percentages.

Carbon ratio theory: The theory that in any area the gravity of oil varies inversely as the carbon ratio of the coal. As temperatures and pressures increase, the percentage of the fixed carbon in coal increases, the rank of the coals rises and the oils become lighter. Increase in metamorphic processes results in elimination of volatile constituents from coal but increases the lighter and more volatile hydrocarbons in oil.

Caving: Caving of rock fragments from the wall of the borehole.

Cuttings: The fragmental rock samples broken or torn from the penetrated rock during the course of drilling.

Fixed Carbon: In the case of coal, coke, and bituminous materials, the solid residue other than ash, obtained by destructive distillation, determined by definite prescribed methods (A.S.T.M. D121-30).

Mud: Various fluids and emulsions, collectively termed, used in rotary drilling.

Organic metamorphism: In order to avoid confusion with the definition of metamorphism accepted by hard-rock petrologists, the term organic metamorphism is suggested. This alteration process of organic material, including pollen-grains and spores, alters under much less severe conditions than most inorganic material.

Proximate analyses: In the case of coal and coke, the determination by prescribed methods of moisture, volatile matter, fixed carbon (by difference), and ash (A.S.T.M. D121-30).

Psilate (smooth): A pollen-grain or spore sculpture type without sculpture elements. Depressions are absent or smaller than one micron.

Rank: Describes the stage of coalification attained by a given coal.

Spore: An asexual reproductive structure, commonly unicellular and usually produced in sporangia.

Trilete: Possessing a triradiate tetrad scar.

Vitrinite: The major maceral, or micropetrological unit of vitrain. It occurs as microscopic lenticels in some coals and is of such consistency as to suggest that it is of the same nature as vitrain.

Volatile matter: Those products, exclusive of moisture, given off by a material as gas and vapor, determined by definite prescribed methods, which may vary according to the nature of the material. In the case of coal and coke the standard methods are A.S.T.M. designation, D 271 and A.S.T.M. D 121-30.

PREVIOUS INVESTIGATIONS

1. COAL INVESTIGATIONS

In his volume on the geology of Pennsylvania, Rogers (1858) noted a consistent regional change in character of the Pennsylvanian coals in the Appalachian basin. The volatile bituminous coals in the slightly deformed rocks of the Pittsburgh coal district were shown to grade eastward into anthracite coals in the strongly faulted and folded rocks of eastern Pennsylvania. Rogers (1865) later demonstrated that a correlation existed between coal composition, oil and gas occurrences, and the degree of metamorphism of the associated strata. He was the first to recognize the "carbon ratio" theory, later elaborated upon by David White (1915).

In the first formal statement of the theory, White in 1915 described the relationship between the limits of known commercial oil and gas occurrences and regional variations in the intensity of rock metamorphism as indicated by the percentage of "fixed carbon" in the coals. In addition, he suggested that the carbon ratio value marking the "deadline" for oil occurrences probably lay between 66 and 75 percent of fixed carbon (White, 1925). White stressed the importance of dynamic thrust as the cause of coal metamorphism, pointing out that normal faults and folds might serve to diminish locally the intensity of metamorphism. He further suggested that differences in the plant constituents of which the coals were made might account for wide differences in carbon ratio values despite an identical metamorphic history of the associated strata. The last suggestion is still valid today.

Since White's original paper, many attempts have been made to evaluate the carbon ratio theory, particularly in Oklahoma (Fuller, 1920), Arkansas Ozarks (Croneis, 1927), north Texas (Fuller, 1919), West Virginia (Reger, 1921; White, 1925), southwest Virginia (Eby, 1923), Rocky Mountains (Dobbin, 1929), western Canada (Jones, 1928; Hume, 1927), northern Alabama (Semmes, 1921), New Mexico (Storm, 1924), Illinois (Moulton, 1925), Kentucky (Russel, 1925), eastern Kentucky (McFarlan, 1926; White, 1925), and Pennsylvania, Maryland, Virginia, and eastern Ohio (White, 1925). One of the best surveys of the history and development of the carbon ratio theory was prepared by Thom (1934). Several causes of variation of individual coal analyses are as follows:

(a) use of nonstandard coal sampling methods
(b) nonstandard analytical work,
(c) differences in coal-forming vegetation at time of burial,
(d) use of "dry, ash free" as compared with "as received, ash free" forms of analyses (Thom, 1934).

If coal carbon ratios are used, these factors must be considered very carefully.

Now standard procedures are common in North America and Europe. The low rank coals are classified (A.S.T.M. = American Society of Testing Materials) according to their calorific values (or British Thermal Units), the higher rank coals are classified according to their fixed carbon or volatile matter (figure 1).

In order to eliminate the effect of the difference in vegetational matter on either the fixed carbon values or the volatile matter and therefore on the rank determination of the coal, M. and R. Teichmüller (1958), M. Teichmüller (1958) and Ammosov (1961) are analyzing the vitrinite of the coal which is a more precise technique than bulk coal analyses.

A recent paper by M. and R. Teichmüller (1958) discusses the carbon ratio theory in general, the relationships between the chemical and physical characteristics of the coals, and the occurrence of oil and gas in Lower Saxony, West Germany. The study indicates that the degree of coal metamorphism in an area can be estimated by reflected- light observations of the vitrinite of the coal. M. and R. Teichmüller (1958) state that all oil fields in the Lower Saxony basin are restricted to the edge of the basin, which contains low-grade coal, whereas in the highly carbonized center of the basin, only gas has been encountered.

Recent papers from Russia and Germany indicate that carbon ratios of coals are being used to evaluate oil potentialities in several areas. A paper by Vyshermirskii (1958) illustrates how the Russians are utilizing the carbon ratios of coals to block out areas in Siberia which are favorable for oil exploration. Vyshermirskii (1958) notes that there is a direct relationship between the carbon ratios of coals and the degree of metamorphism of the sediments in all regions studied. According to Vyshermirskii, this method of forecasting oil bearing capability has long been known, but "in practice of the Soviet oil men, however, this method did not gain wide use. This is probably due to the fact that up until recently the main volume of oil exploration was conducted in regions almost devoid of coal outcrops."

As a result of extensive work by Ammosov (1961), it has been determined that all main oil fields in the U.S.S.R. are associated with sediments in which the coal carbon ratio is not higher than 60, with isolated oil fields in the fixed carbon range 60—65 (fig. 1). Since in many basins of the world coals are either rare or absent, the carbon ratio method was of limited use

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1 Calorific value of the moist, mineral-matter-free coal.
2 On a ash-free-dry basis.

Fig. 1. Relationship between rank of plant remains and the occurrence of commercial oil and/or gas in reservoirs.

* ECONOMIC COMMISSION FOR EUROPE 1956, AND UNITED NATIONS

** FIXED CARBON

*** EXPRESSED IN 10^{-3} MICRO AMPERES

\+ CORRELATION WITH U.S.A. CLASSIFICATION

BY K. PATTEISKY 1953

for determining the incipient metamorphism of the sediments. World-wide palynological studies have, in general, shown relative abundant pollen-grains and/or spores and other plant matter in the sediments of many basins. If pollen-grains or spores could be used in sediments to express the level of organic metamorphism, then basins with sporadic coal occurrences or devoid of coals could be investigated.

2. CHANGES IN THE EXINES OF POLLEN-GRAINS AND SPORES DURING CARBONIZATION

The pollen-grains and spores in coal beds, studied both isolated by maceration and by thin section, change in color with increasing coal rank from yellow over light brown to dark brown and finally to black. In the range brown coal to high-volatile B bituminous coal pollen-grains and/or spores are in general yellow. From high-volatile A bituminous coal to low-volatile bituminous coal pollen-grains and/or spores change in color from yellow over brown to black. In the past these qualitative changes of pollen-grains and spores have been reported by several authors (Bode, 1928; Potonie, 1930; Kirchheimer, 1934; Ergol'skaya, 1939, 1947; Schopf. 1948; Böttcher and M. and R. Teichmüller, 1949; M. and R. Teichmüller, 1950; Val'ts, 1952; Bogolyubova, 1956; M. Teichmüller, 1958; Ammosov, 1961; Wilson, 1961; Schopf, 1962). No spore exines have been found in coals ranking higher than "Magerkohlen" (equivalent to semi-anthracite, figure 1) (Bode, 1928; Potonie, 1930). This has been caused by chemical changes which transformed pollen-grains and spores into totally carbonized particles. Microspores on polished sections from coals with vitrinite containing 20 to 22 percent volatile matter become invisible (Teichmüller, M., 1958). It is of interest in connection with the observations of M. Teichmüller to note that a similar change of pollen-grains and spores has been noticed by Schopf (1948) in thin sections in coals within the range of 23 to 25 percent volatile matter. The pollen-grains and spores from other sedimentary rocks, such as shales and sandstones, change in similar color sequence as is observed in coals, indicating their low-grade metamorphism. It seems that strata with levels of organic metamorphism equivalent to semi-anthracite, anthracite and meta-anthracite are unsuitable for palynological time correlation or ecological investigations. Pollen-grains and spores in these levels are completely carbonized. Sediments with levels of organic metamorphism equivalent to the lower part of medium volatile bituminous coal and of low volatile bituminous coal may contain pollen-grains and spores

less suitable for palynological studies, because of their dark brown to black color. Early recognition of the above facts may help the palynologist in the selection of favorable areas for palynological studies.

3. POSSIBLE CAUSE OF CHANGES IN THE LIGHT ABSORPTION OF POLLEN-GRAIN AND SPORE EXINES DURING CARBONIZATION

It is interesting to speculate why light-reflection and light-absorption values of plant material (respectively by reflected and transmitted light) change during the coalification process.

The reflection index and the absorption coefficient of coals depend on the chemical structure of the coal. Also the reflection capability of vitrinite under reflected light (van Krevelen, 1953; H. Stach and M. Teichmüller, 1953) and the absorption of transmitted light by pollen-grains and spores (Gutjahr: this study) depend on the chemical structure of the constituent under consideration. The absorption capability of plant material for light, according to the electromagnetic light theory, depends on the number of electrons with free oscillation, present in the plant material. The oscillation of transmitted light are dampened by the oscillations of the electrons of the plant material and cause light absorption (M. and R. Teichmüller, 1954).

The following statements are taken from R. W. Terhune (1964, p. 39).

"When an ordinary light beam passes from air into a transparent material the most obvious phenomenon is that the light beam slows down. The amount of bending of the light beam, according to geometric optics, depends on the ratio between the index of refraction of air and that of the transparent material. There is therefore an interaction between light and matter. In physical optics, this interaction is viewed as occurring between the oscillating electric field of the light beam and the electric charges in the atoms or molecules in the material. When the beam passes through a material, the oscillating electric field moves the electron cloud surrounding the positively charged nucleus first in one direction, then in the other. The net effect is that each particle becomes a miniature oscillating dipole radiating energy at precisely the same frequency as the incoming wave.

Each dipole acts as a separate source and the scattered radiation from them is called Rayleigh scattering. In the forward direction, both the incident and the re-radiated fields are present. The total field is, therefore, the vector sum of their amplitudes. Although the two fields are phase coherent, the re-radiated field is delayed 90 degrees in time phase from the incident field. The result of this vector addition is a wave with lower velocity than that of the incident wave."

CAUSES OF CARBONIZATION

1. INTRODUCTION

From a physical-chemical point of view, the coalification process can be subdivided into three stages (Huck and Karweil, 1962). In the beginning of the coalification process complex chemical reactions are caused by fungi and bacteria. In the second stage (the formation from peat to high volatile bituminous coals, figure 1) the bacterial and chemical changes decrease in favor of a mechanical compression of sediments, which causes expulsion of water. The last stage (the formation from high volatile bituminous coal to anthracite, figure 1) is a combination of physical and chemical changes. The chemical metamorphism of coals is characterized by an increasing aromatization of humic substances. During this process molecule groups of nonaromatic character rich in hydrogen and oxygen are released. At the same time the remaining molecules of aromatic character are growing together (M. and R. Teichmüller, 1954). Among the factors causing carbonization reported in literature are temperature, time, pressure, and radio-activity. If the main cause of carbonization can be found, severely carbonized zones can be predicted. A knowledge of the cause of carbonization therefore is of importance.

2. TEMPERATURE

Recent *Quercus robur* (oak) pollen-grains on a glass slide have been subjected to a temperature controlled hot plate in the open air. The author does not suggest that a similar process will occur in the subsurface of the earth but warns that pollen-grains and spores should not be heated above 100 °C if carbonization measurements are being conducted. The *Quercus* pollen-grains have been heated at 100 °C, 150 °C, and 200 °C for approximately one week (figures 2 and 3). Slight changes in carbonization could be observed if *Quercus robur* pollen-grains were heated at 100 °C for a week. However, if they were heated at 150 °C and 200 °C, important carbonization changes could be observed (figure 2 and 3). A plot of mean carbonization (light absorption) values against time (figure 2) show that for each of the temperatures the rate of carbonization change decreases rapidly and that after some 50 hours of temperature exposure a level is reached and only slight or no further carbonization changes will occur. If cuttings samples, cores, or sidewall cores from wells or Recent samples are used for pollen-grain and spore carbonization studies, temperatures of less than 100 °C are recommended if samples are dried, because temperatures of 100 °C, 150 °C and 200 °C carbonize pollen-grains and spores in a relatively short time interval. This experiment shows that temperature can carbonize pollen-grains.

An example of slight temperature exposure in nature during a considerable time interval is the Lower Carboniferous brown coal in the basin of Moscow. This deposit has never been buried deeply and it has been exposed to low temperatures only. Examples of high temperature exposure in nature are the severe carbonization zones near small or large igneous bodies. Examples are the Pliocene brown coals of Palembang, Sumatra, Indonesia (Mukherjee, 1955), which by contact metamorphism are partly changed into anthracite, and the organic metamorphic zones around the Bramscher massif, German Federal Republic (M. and R. Teichmüller, 1958). These and other studies suggest that temperature is the predominant cause of carbonization.
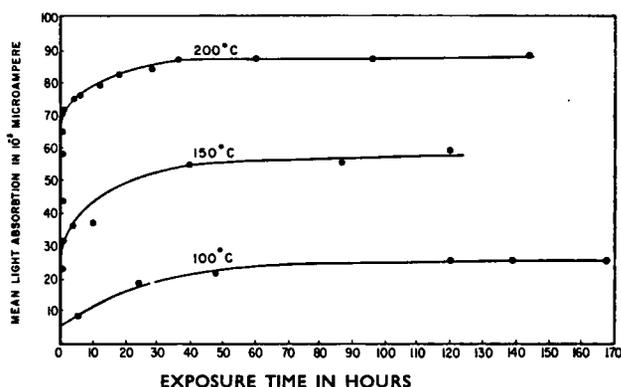
Fig. 2. Mean light absorption values of *Quercus robur* pollen-grains as a function of exposure time and temperature.

3. TIME

The effect of exposure time of low temperature on pollen-grains and spores is not important. The experiment discussed in Section 2 of Chapter II shows this clearly. One of the best examples in nature of the effect of exposure time of lower temperature on organic matter is the Lower Carboniferous brown coal of Moscow. Apparently time alone could not carbonize this carbonaceous material beyond the brown coal rank (M. and R. Teichmüller, 1954). The effect of exposure time of higher temperatures on pollen-grains is of importance. Slight carbonization changes in *Quercus robur* (oak pollen-grains) could be obtained at 100 °C for a week (figure 2), but drastic carbonization changes were observed at higher temperatures (viz. 150 °C and 200 °C) in a relatively short time interval. Huck and Karweil (1955) have published a diagram (figure 4) showing the relationship between carbonization, time, and temperature. The diagram indicates that impor-

* APPROXIMATE

Fig. 3. Light absorption data sheet of *Quercus robur* (oak) showing the relation between light absorption, temperature and exposure time.

tant changes in carbonization can be obtained if carbonaceous matter is exposed longer at relatively higher temperatures.

4, PRESSURE

White (1925) emphasized the importance of "dynamic thrust" as the cause of coal metamorphism. However, Lesley (1879) had earlier proposed that the regional change in the rank of coal in Pennsylvania was due to the higher rank coal having been buried more deeply. He pointed out that the difference in the amount of the original overburden could easily explain why "disturbed" Belgian coals are not anthracites and why Arkansas anthracites are not disturbed.

M. and R. Teichmüller (1954) mentioned a brown coal in a folded belt in Pakistan which was greatly deformed by dynamic thrusting but was only slightly altered in rank.

The western part of the Prokop'evsk-Kisel area (Kuznetsk basin, Russia) is characterized by steeply folded and faulted coal measures (I. I. Ammosov, 1961). These coals are less coalified than the coals in the less disturbed parts of the basin.

In the Westphalian and the French-Belgian coal fields, coal beds are richer in volatile matter (less carbonized) on the highly folded and faulted south side of the coal basin than they are on the north side where the strata are much less disturbed (Reeves, 1928). The above examples show that pressure alone apparently could not increase the rank of the coal.

Various coal samples were subjected (Hoffman, 1936) for four days to a pressure of 18,000 kg per square centimeter. Hoffman noted an average decrease in volatile matter by 1.5 %, indicating very slight carbonization.

According to Hoffman (1936) and Bergius (1913) coalification by pressure is successful only if accompanied by relative high temperature. This was also observed by Gropp and Bode (1932) who noticed a

decrease of volatile matter of "immature" coals when subjected to 300 °C and 1800 atm. Gropp and Bode (1932) considered the high temperatures in the synclines of deep basins as the main factor in the carbonization of coal, while pressure only hinders the formation of carbonization products.

Trifonow and Toschew (1940) compressed nine different coal samples by applying a pressure of 10,000 kg per square centimeter and only slight increase of total volatile matter was noticed.

From a thermodynamic viewpoint (Fuchs, 1941), rising pressure in a condensed heterogeneous system will cause phenomena making for decreasing volume, e.g., boiling points will be raised, melting points will be lowered and simple polymerization will be favored. Hence only small changes of proximate analysis, but no significant change of ultimate composition or rank of coal may result from compression.

Also, van Krevelen (1957) mentioned that static pressure has no effect on the chemical properties of the coal since it is inconsistent with thermodynamics. Compression may, conceivably, cause a rise of temperature. Fuchs (1941) calculated that, assuming a maximum compression of 10,000 kg per square centimeter, a starting temperature of 25 °C, a specific heat of 0,3 cal per gram coal, a volume expansivity $= a$ of 0.2 × 10^{-4} and a specific gravity of 1.3, a temperature increase of 3.5 °C could be expected which is obviously negligible for coalification (carbonization).

Fuchs (1953) and Huck and Karweil (1953) have stated that pressure has a great influence on structural alteration of sediments but that pressure alone cannot change the chemical nature of the coals. Huck, Karweil, and Pateisky (1962) used a heatable pressure devise in order to study the coalification process. They concluded that static pressure delays coalification and does not accelerate the coalification process as is often assumed.
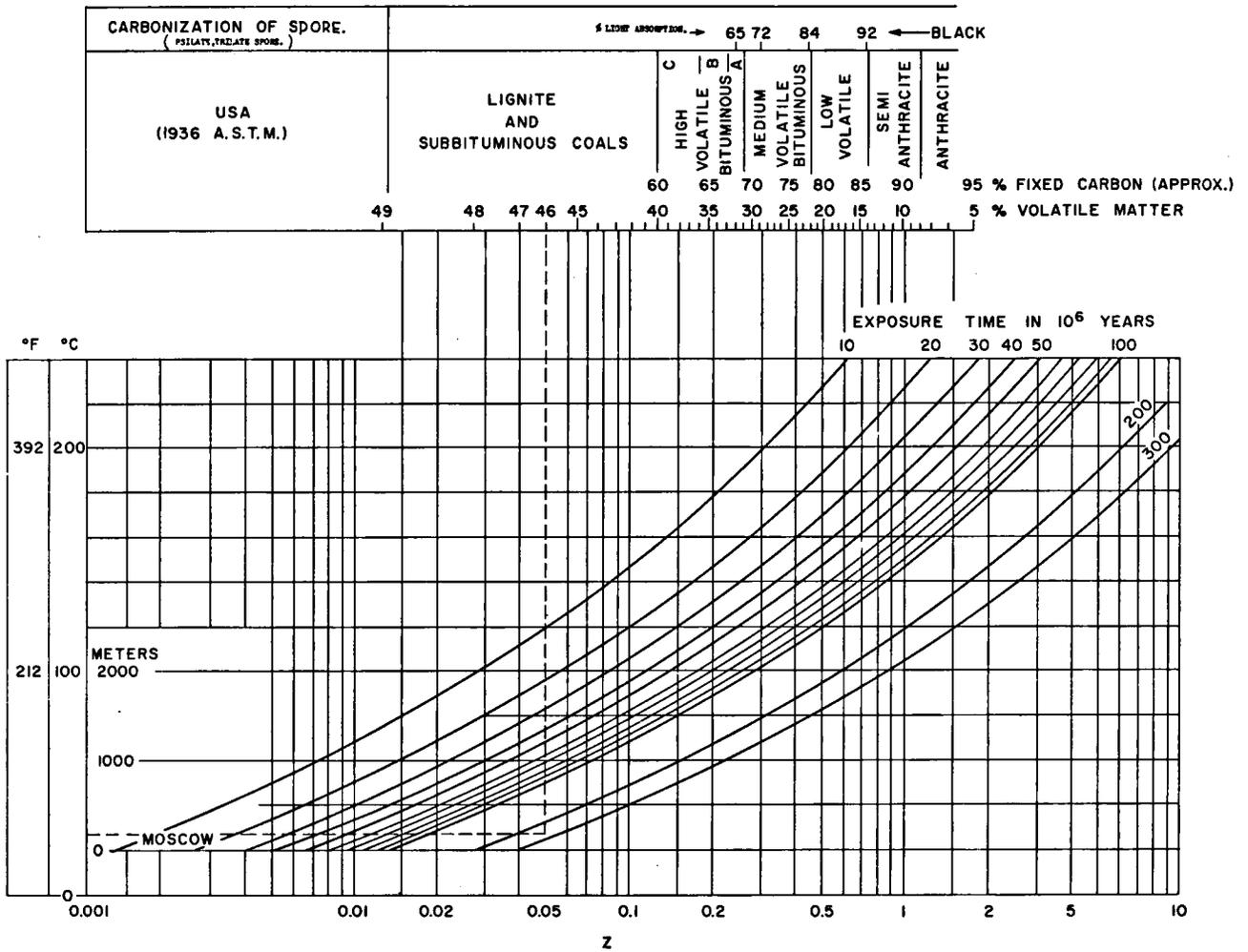
Fig. 4. Relation between carbonization, temperature and exposure time (Karweil 1955, with additions by the author).

Concerning the dynamic pressure in relation to coalification, the same authors mentioned that the influence of dynamic pressure in nature cannot be ignored, but that it is rarely of importance.

E. Lewis (1936) found that "cokes" formed from a coal, when heated under diminished pressure, lie on straight lines (RS, TM, and AB of figure 5a). When, however, coals were heated under pressure, a series of "cokes" was produced (see curved lines JKP and NOP of figure 5b) whose compositions more nearly approximated to the compositions of natural coals (figure 5b). The composition of natural coals is indicated in figure 5a and 5b as a gray band, the so-called coal band.

Regarding the change in composition of a coal under influence of temperature and pressure, Lewis' (1936) study shows that the effect of pressure produces residues that are closer to the coal band than the residues obtained under the action of heat alone.

Carbonization studies of a lignite in pressure vessels by J. Rogers, R. P. Suggate and J. O. Elphick (1962) show (figure 6) that at 1000 atmospheres and at 170 °C

and above there is an initial rapid change. whereas up to 100 °C the change in calorific value or in coalification is slow in terms of time of these experiments. It is interesting to note that at 170 °C there were greater changes in calorific value or carbonization at 10 atmospheres pressure than at 1000 atmospheres. This observation is in agreement with the observation of Huck, Karweil and Pateisky (1962) viz. that static pressure retards coalification. Rogers, Suggate and Elphick (1962) furthermore found that there is a marked point of inflexion at about 170 °C in the the calorific value versus termperature (figure 6), obtained at 1000 atmospheres pressure over a period of seven days, the inflexion point at 10 atmospheres and seven days however, is only at about 120 °C. It seems that carbonization is slow at low temperatures until a particular temperature (threshold T) is obtained after which rank increases more rapidly with increasing temperatures. The higher the pressure, the higher this threshold temperature will be (Rogers, Suggate, Elphick, 1962).
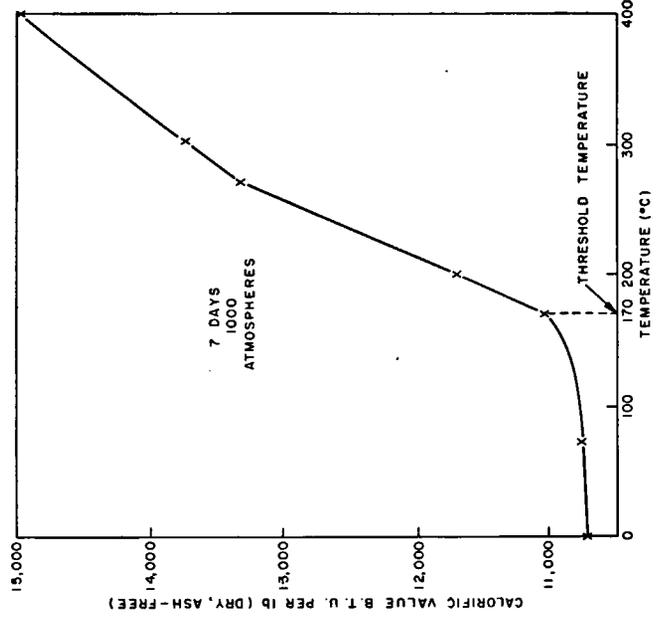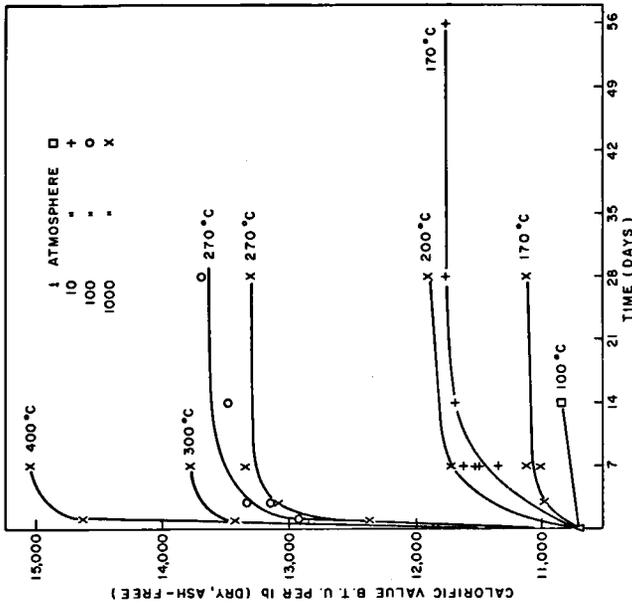
Fig. 6. The effect of temperature and pressure on coalification (Rogers, Suggate, Elphick and Ross, 1962).
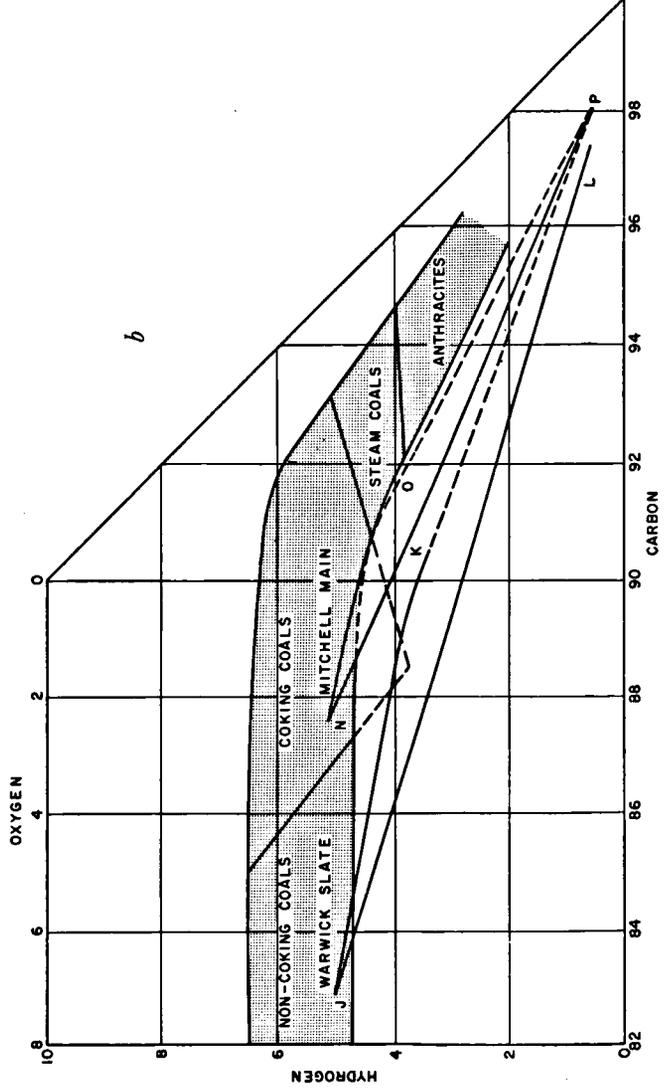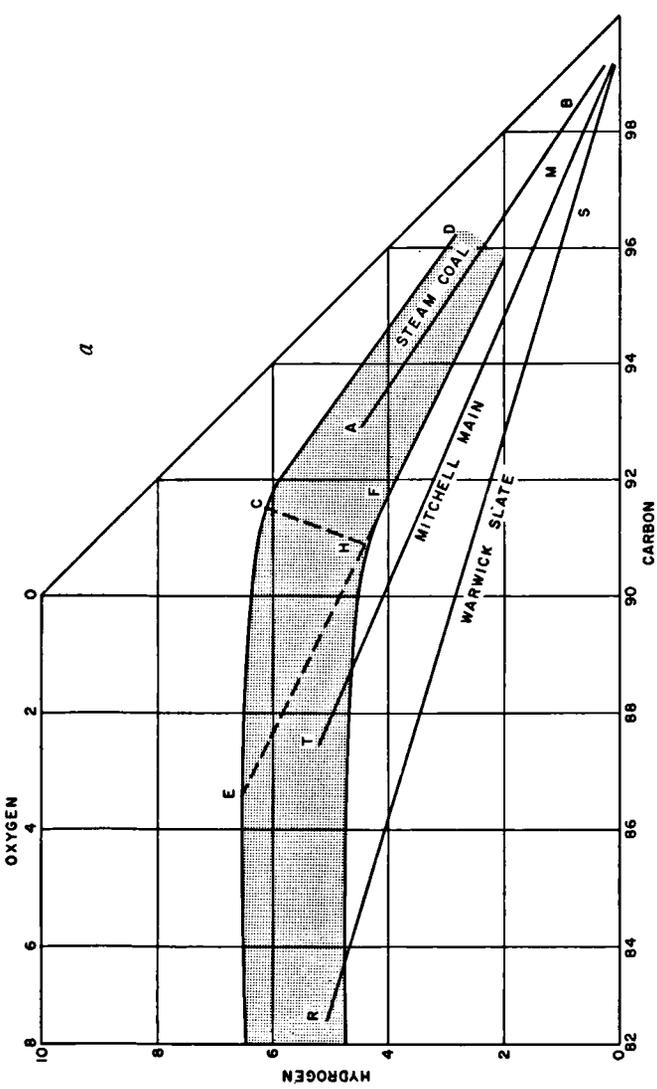
Fig. 5. a) Effect of heat under diminished pressure on coal (Lewis 1936).
b) Effect of heat at 300 lbs. pressure on coal (Lewis 1936).

EQUIPMENT FOR CARBONIZATION MEASUREMENTS OF POLLEN-GRAINS, SPORES, AND OTHER PLANT FRAGMENTS

1 DESCRIPTION OF EQUIPMENT

The microscopes listed below will not be described in detail, since they are well-known standard microscopes. They must have features normally required for palynological studies. These would include mechanical stage, several low-power dry objectives, an oil immersion objective, and an ocular scale or similar device to provide a visual reference point.

I. *Standard equipment*

A. *Carl Zeiss Standard Microscope* (figure 7) — with inclined binocular tubes and a vertical phototube with 8 × or 10 × ocular.

B. *Carl Zeiss photographic equipment* (figure 7)
1. A basic body FL II with 35 mm camera back. The camera back is not necessary for carbonization studies.
2. A photocell in housing which is carried on the basic body FL II or on a special extension tube (figs. 8b and 10). The photocell in housing (Zeiss catalog no. 47-42-10) is connected to a separate four-stage amplifier (Zeiss catalog no. 47-42-01).

The photocell is a phototube with a characteristic linear curve and photoelectric response that is stable over a long period of use. It should be noted, by comparison with phototube, that the response of chemical photoelectric cells (selenium, etc.) is unstable, exhibiting a gradual deterioration with use.

C. *Carl Zeiss photomicroscope* (figures 8b an 10) — with a special extension tube which carries the photocell in housing.

D. *Other standard microscopes* — with inclined binocular tubes and vertical phototube.

It is probable that the photographic equipment can be adapted to the vertical phototube of any microscope because tube diameters are reasonably standardized.

II. *Special equipment and modifications*

A. *Field restrictor* (figures 8a and 8c)
This is the most important item of the special equipment. Care must be exercised in making the field restrictor so as to have no rough surfaces in the holes
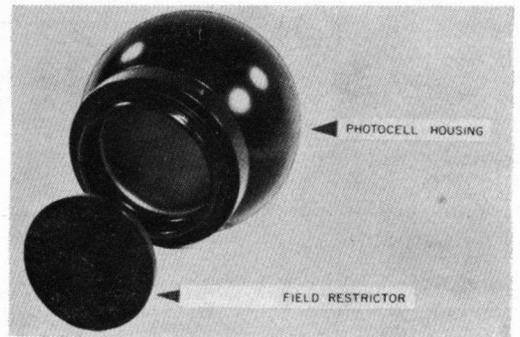
Fig. 8a. Field restrictor and photocell housing.
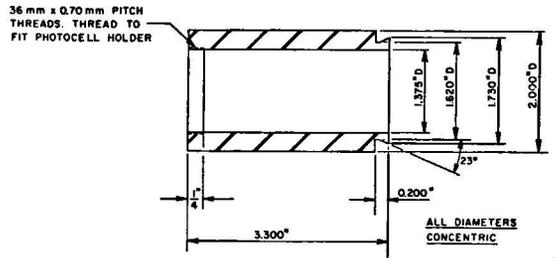
Fig. 8b. Extension tube for photomicroscope (after Prescott).

Fig. 7. Standard microscope equipped for light absorption measurements.

Fig. 8c. Field restrictor (after Prescott).
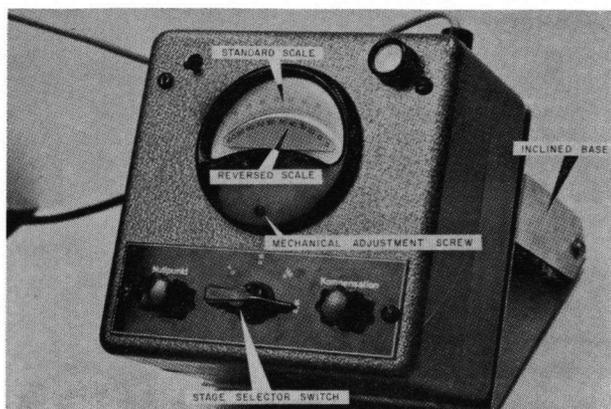
Fig. 9.   Four-stage amplifier equipped with reversed scale for-light absorption values.

Fig. 10.   Photomicroscope equipped for light-absorption measurements.

and to soft-radius both holes. This is done to minimize any edge diffraction. All parts are thoroughly black-anodized before assembly. The restrictor should be fitted to its individual photocell in housing.

B. *Reversed scale* (figure 9)
The reversed scale of the four-stage amplifier is used instead of the standard scale supplied. This is done to provide numerical values that increase with increasing light absorption.

C. *Inclined base* (figure 9)
The inclined base of the four-stage amplifier permits more convenient and rapid reading of the scale; it is simple to construct and therefore specifications are not given.

D. *Collar* (figure 7)
A collar with set screw was constructed to fit around the vertical phototube below the attachment fitting of the basic body FL II. This permits exact repositioning of equipment. Because the collar may not be necessary, and since it is simple to construct, specifications are not given.

E. *Special extension tube* (figures 8b and 10)
If a Carl Zeiss photomicroscope is used, a special extension tube is added that carries the photocell in housing, which is connected to a four-stage amplifier.

F. *Control center* (figure 7)
The control center consolidates the Sola transformer (described below) and the variable voltage transformer of the microscope lamp and furnishes the necessary electrical grounding.
The Sola transformer is a standard item which is not normally needed for palynological studies but is used

here to prevent errors induced by voltage fluctuations. The Sola standard sinusoidal (C.V.S.) constant-voltage transformer with a Sola adaptor Kit for input plug and output jack is used for maximum electric stability. This type of constant-voltage transformer was selected to minimize the possible effect of harmonics on the amplifiers.
The additional equipment does not effect the use of the microscope for routine palynological correlation studies, nor does it affect the use of the Carl Zeiss photomicroscope for normal photography. The modification to the photoelectric cell of the Zeiss attachment camera as used with the standard Zeiss microscope does not seriously affect the equipment for photographic use. The restriction of light by the field restrictor to the photocell makes it necessary to determine exposure values for meter readings.
As previously noted the equipment described here consists largely of Zeiss equipment. The total equipments has performed satisfactorily and is reasonably inexpensive. Other brands of equipment could probably be used. However, for comparison of data, the photoelectric cell characteristic should be similar to those of the Zeiss phototube, and the amplifier should be at least of equal accuracy and sensitivity.
The equipment used in the two different modifications described above is identical in many respects. The field restrictor, the photocell, the modified four-stage amplifier, and the constant-voltage transformer are the same and are interchangeable. The differences are in the adaption of the equipment to different microscopes.

PROCEDURES

1. STANDARD CHEMICAL PREPARATION TECHNIQUE

Spores and pollen-grains are affected to a greater or lesser extent by certain chemicals, and a standard treatment insures that variables will not be introduced by laboratory preparation methods.

Mechanical treatments will not affect the absorption characteristics of pollen-grains and spores. However, prolonged ultrasonic vibrations may crack or fragment specimens which then cannot be measured accurately.

The use of oxidants have been avoided (nitric acid, Schultz's solution, acetolysis, etc.) in the following standard preparation procedure (for calcareous shales, Steps 1-14, for shale, dirty sandstones, and silt, Steps 3-14). The following steps have been used in this investigation:

(1) pulverize sample, and add a solution of 20 percent hydrochloric acid; boil four minutes,

(2) centrifuge, decant, and add water to residue; centrifuge again,

(3) add hydrofluoric acid (52 percent) to residue; boil five minutes,

(4) centrifuge, decant, and add water to residue; centrifuge again,

(5) boil residue with hydrochloric acid (20 percent) for five minutes,

(6) centrifuge, decant, and add water to residue; centrifuge again,

(7) boil residue for 30 seconds in potassium hydroxide (5 percent),

(8) centrifuge, decant, and add water to residue; centrifuge again,

(9) put residue in aqueous suspension in test tube,

(10) insert in an ultrasonic generator,

(11) centrifuge and decant, normal or short centrifuging,

(12) add alcohol, shake, and centrifuge,

(13) separate with heavy liquid using bromoform — alcohol solution (s.g. 2.0),

(14) put float from Step (13) in vial.

It is probable that slight modification of the above standard treatment may yield comparable results. However, for greater confidence in comparing carbonization data obtained in different laboratories, it is suggested that the above preparation procedures be adopted.

2. SELECTION OF STANDARD POLLEN-GRAIN OR SPORE

One of the most important considerations in undertaking light-absorption value measurements is the selection of the type or "species" of the pollen-grain or spore to be measured. It must be recognized that different types have different light-absorption value because of original differences in wall thicknesses and possible original differences in chemical composition. At any given degree of low-grade metamorphism, there will be pollen-grains and spores with different light-absorption values. With increasing metamorphism these absorption differences become less pronounced. It is important to select a type that will give low absorption values when slightly carbonized. If a type with high initial light-absorption values is chosen, the measurable effect of light-absorption alteration is drastically reduced (figure 11).

The standard type should be relatively large, because an area of the exine should be selected which is free of aperatures, occasional folds, etc. The exine should be reasonably uniform in thickness, relatively free o ornamentation, preferably smooth (psilate), and generally without cracks, folds, or perforations. The type should be easily recognizable and distinctive. The vertical range and relative abundance of the types available for selection should be considered. It
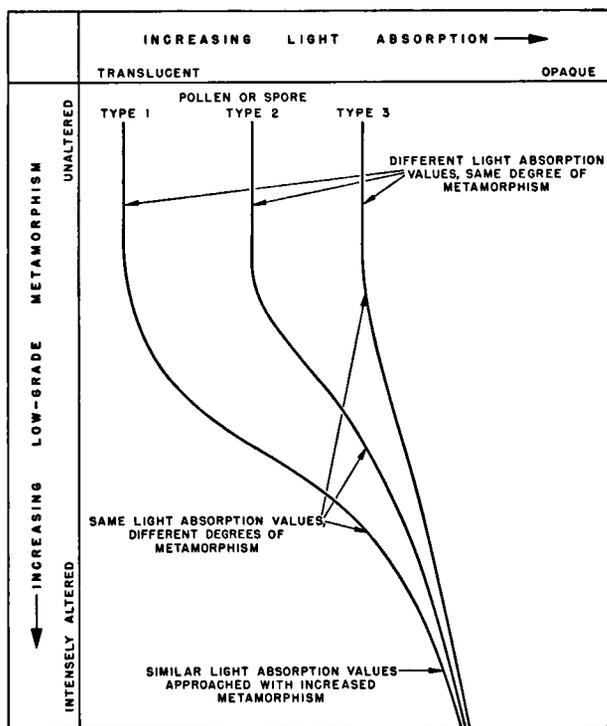
Fig. 11. A schematic comparison of the effect of increasing low-grade metamorphism on the light absorption of three different pollen-grains or spores.

is preferable that the type chosen be relatively abundant throughout the strata to be studied.

If the chosen type becomes relatively rare or disappears in the samples investigated, another type may be selected. The light-absorption values of these different types can be equated in zones in which both occur.

3. PRE-OPERATIONAL PROCEDURES

a. *Four-stage amplifier adjustments* (figure 9)

(a) The indicator of the microampere scale (standard scale) may have to be mechanically adjusted to *0* by adjusting the screw near the base of the dial. The stage selector switch (middle knob) must be on *0* during this procedure. Usually this must be done when the equipment is received from the supplier and at regular intervals, but it is not a part of the operational procedure.

The transformer and the four-stage amplifier are switched on to warm up. It should be noted that longer life and stability can be expected if the four-stage amplifier is turned on Monday morning and is turned off only for the week-end.

(b) The four-stage amplifier is zeroed with the "Nullpunkt" knob on the *0* of the standard scale with the stage selector switch on *1*.

The amplifier is compensated with the "Kompensation" knob on the *0* of the standard scale with the stage selector switch on *1000*. In both adjustments no light has passed to the photocell. The zero and compensation should be checked at regular intervals each day.

b. *Location of the sensitive spot*

The field restrictor confines the field to be measured and defines a sensitive spot or area, approximately six microns in diameter (with the oculars described), with an oil immersion objective. The exact position of this spot is slightly different for each microscope and restrictor equipped photocell.

The sensitive spot is located by the procedure outlined below. The amplifier is zeroed and compensated, and a small ($\sim$ 5 micron) opaque black particle is located under oil immersion in a field that is relatively free of other debris.

The beam-splitting slide is pulled to permit light to the photocell, and the stage selector switch is set on *1000*. The black particle is moved almost out of view, and the lower diaphragm of the microscope is adjusted to obtain *0* light absorption on the reversed scale.

The black particle is moved across the field of view by multiple traverses until the maximum light absorption reading is obtained. The position of the sensitive spot in the field is the position of the black particle at this maximum light absorption reading (maximum reading on reversed scale). The position of the sensitive spot in the field is located for future reference on the scale in the ocular. The accurate location of the sensitive spot in the field is fundamental and should be checked occasionally. Once the location of the sensitive spot is established, neither the field

restrictor in housing nor the ocular with reference scale should be moved.

It is suggested that the ocular then be fixed (taped or more permanently fixed) to the body of the inclined ocular tube, because if it and the included scale are rotated after positioning, the reference point to the sensitive spot will be lost. The beam-splitting slide can be moved to shut light from the photocell, and measurement of pollen-grains and spores can be started.

4. OPERATIONAL PROCEDURES

a. *Operational procedures for standard Zeiss microscope with modified photographic equipment*

(a) The pollen-grain or the spore to be measured under oil immersion is offset so that the sensitive spot is located in a fully illuminated vacant area, free of debris.

(b) The beam-splitting slide of basic body FL II is pulled to position 3 (completely out) to permit light on the photocell. The lower microscope diaphragm is adjusted so that the light produces a reading of *0* light absorption on the reversed scale of the four-stage amplifier when set on its most sensitive scale (*1000* on the stage selector switch).

The technique of adjusting the light available to the photocell to produce a scale reading of *0* light absorption (zero on reversed scale) before each pollen-grain and spore light-absorption measurement, eliminates several variables (e.g., differential thickness of microscope slide, cover glasses and variations in mounting media). The light source is kept constant by the Sola constant voltage transformer throughout these operations.

(c) The pollen-grain or the spore is moved so that the sensitive spot is located in the area to be measured. Folds, cracks, thicker and thinner areas in the exine, and debris above and below the pollen-grains or the spores must be avoided.

(d) The light absorption value is read on the reversed scale of the four-stage amplifier meter and is recorded.

(e) The beam-splitting slide is returned to position 1 or 2 so that no light is cast on the photocell. Steps (a) through (e) are repeated for each measurement. The amplifier remains on during operations.

b. *Operational procedures for Zeiss photomicroscope with modified equipment*

The light is excluded from the photocell at positions 1, 2 and 3 of the built-in beam-splitting slide. Normally position 2 is used for observations, and position 3 which masks the area to a 35-mm frame size with cross hairs, is used to locate the sensitive spot.

(a) Same as Step (a) above, except that for consistent magnification the ocular of the microscope is always set on the same magnification factor.

(b) Same as Step (b) above, except for beam-splitting slide position, which is pulled to position 4 to pass light vertically to the photocell.

(c) Same as Step (c) above, except that the beam-

splitting slide must be returned to position 3 for observation and moved back to position 4 for measurement.
(d) Same as Step (d) above.
(e) The beam-splitting slide is returned to position 1,

2 or 3 so that no light is passing through the photo-cell.
Steps (a) through (e) are repeated for each measurement, with proper position of the beam-splitting slide. The amplifier remains on during operations.

CHAPTER V

RECORDING, PRESENTATION AND EVALUATION OF DATA

1. RECORDING AND PRESENTATION OF DATA

The numerical values read on the reversed scale of the four-stage amplifier are termed light-absorption values expressed in millimicroamperes. Several methods can be devised to record and present graphically these absorption values of measured specimens of the same type.

The absorption values of individual specimens are recorded as they are encountered during traverses of the slide. It is desirable to measure approximately 40 specimens of the same type per sample. With increasing low-grade metamorphism less variation in light-absorption occurs, and fewer measurements per sample are sufficient.

The arithmetical mean or the median light-absorption value can be calculated for each sample depending on the distribution of the light-absorption values. To determine the reliability of the arithmetical mean of each sample, the approximate 95 percent confidence interval can be calculated by standard methods.

From these data a *carbonization diagram* is then plotted, with light-absorption values, expressed in millimicro-amperes, as the horizontal scale and depth as the vertical scale (figure 12). For each individual sample a frequency curve is constructed with its base at the sample depth and its height controlled by an arbitrary but uniform vertical scale. These curves have a characteristic "saw blade" pattern. Each point on the curves represents the number of specimens with a given light absorption value. The average light absorption value for each sample is plotted at the base of the frequency curve. These points are connected in succeeding samples to show a line representing the trend of the values called the *carbonization* line. This line is drawn for each well or section and is used to estimate the depth at which certain average light absorption and, thus, carbonization values are encountered.

Carbonization lines for different wells in sections, panel diagrams, etc., can be used to show the carbonization trends in compressed form. In section lines connecting the same light absorption values can be constructed; these are referred to as *isocarbonization lines* (figure 13a). The intervals between succeeding isocarbonization lines, here referred to as a *carbonization zone* (figure 13a), with a specific light absorption range, can be color-coded in sections in order to aid the visual presentation. If carbonization measurements of

an adequate number of wells in a certain area are available, several types of contour maps can be drawn. One type shows the depth of a specific carbonization value (or a specific light absorption value). These contours are termed *isocarbonization contours* (figure 13b). Another type shows the carbonization values of a specific datum, which can be either a geological unit or a specific depth. These contours are termed *carbonization contours* (figure 13c). The type of contour map(s) to be made depends upon the purpose of the investigation. Block diagrams, panel diagrams, etc., can be constructed with either type of contour map.

2. EVALUATION OF DATA

The data obtained by the method discussed in Chapter IV and Chapter V section 1, are numerical expressions of the light absorption of pollen-grains and spores. Empirical data have repeatedly shown that these values change with changing carbonization of coals.
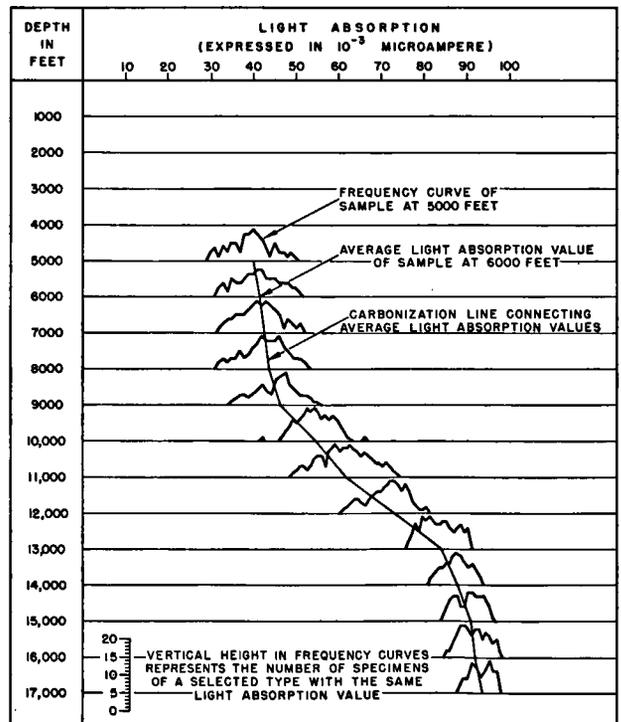
Fig. 12. Schematic carbonization diagram.

As carbonization increases during the coalification process, the spores and pollen-grains become darker and absorb more light. The absorption values of a specific type in succeeding samples in a well are therefore considered to be a measure of the carbonization stage, or "rank", of the strata.

Carbonization is a low-grade metamorphic process
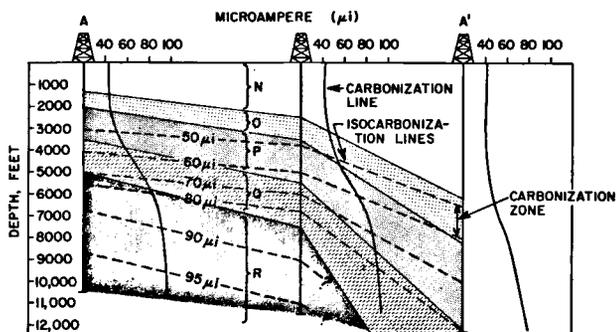
Fig. 13a.   Schematic cross section showing carbonization lines, isocarbonization lines, and carbonization zones.
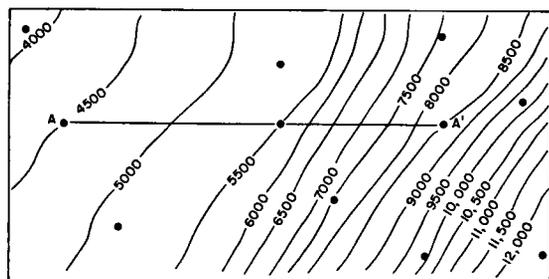
Fig. 13b.   Schematic map with isocarbonization contours showing the depth of a specific carbonization line (65 μi).
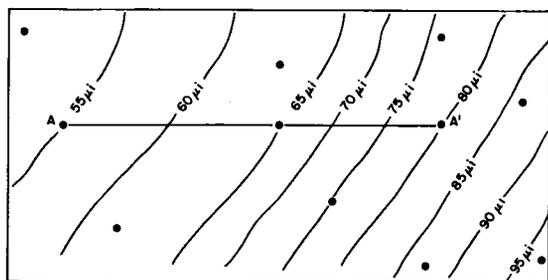
Fig. 13c.   Schematic map with carbonization countours showing the values of a specific datum (P-Q contact).

which alters organic material, and the absorption values are therefore considered as indicators of this organic metamorphic process.

Carbonization begins initially with slightly altered (translucent) plant material and continues toward intensely altered (opaque) plant material. The term "carbonized" has been applied in palynological studies to spores, pollen-grains, and other plant materials with various meanings. In general, the term ha, been used to indicate that either the spores and pollen-grains or the associated plant debris, or both, have been so altered that the isolation and identification of spores and pollen-grains is impossible. In some cases the term has been used when black plant debris is present, with or without spores and pollen-grains. In general most plant material undergoes more rapid carbonization alteration than the associated pollen-grains and spores. These usages of the term carbonized are unfortunate, because unwarranted conclusions may be drawn. According to our usage, virtually all fossil plant material is carbonized to some extent. Such terms as "completely carbonized" or "totally carbonized" are proposed in this study to indicate that pollen-grains, spores and other plant debris are black (opaque).

The contamination of a sample by spores and pollen-grains which have had a different carbonization history may alter the general aspect of that sample. For example, contamination in any manner by Recent pollen-grains and spores which are less carbonized. mens to a sample. Also, caving usually introduces pollen-grains and spores which are less carbonized. Reworking of sediments, on the other hand, usually introduces types that are more carbonized than those which were deposited in an unaltered state at the time the strata were deposited. Furthermore, the pollen-grains and spores introduced by the drilling mud on and in the cores, sidewall cores, or cuttings may be derived from penetrated rocks, from mud additives, or from Recent plants. These mud-introduced pollen-grains and spores may be either more or less carbonized than the pollen-grains and spores from clean rock samples.

In our experience to date these various types of contamination usually do not severely limit carbonization studies. In order to introduce serious errors, the sample must be contaminated by considerable numbers of a type identical with the the standard pollen-grain or spore type measured. Precautions taken in normal routine palynological studies are sufficient for satisfactory carbonization studies.

EXAMPLES OF CARBONIZATION MEASUREMENTS

1. COAL SAMPLES

Spore and plant fragment carbonization measurements of humic coal samples have been made. The coal samples were provided by Dr. M. Teichmüller, Geologisches Landesamt, German Federal Republic, Europe and by Mr. F. E. Walker, Bureau of Mines, Pittsburgh, Pennsylvania, U.S.A. (fig. 14, pp. 000, 000).

Light-absorption measurements of plant fragments in the insoluble residue with a specific gravity of 2.00 and less have been made for humic coal of different rank ranging from lignite to meta-anthracite (figure 15). Although varying in thickness and tissue type, plant fragments can, by their opacity, give an approximate measure of carbonization rank. In the low-rank coals analyzed (lignite to high-volatile B bituminous coal), the plant fragments colors range from yellow to brown to black (the light-absorption values range from 35 to 93 × 10^{-3} microampere). In the range high-volatile A bituminous coal to semi-anthracite, the plant fragments vary from dark brown to black. In the anthracite and meta-anthracite ranks, regardless of the thickness and type of plant fragments, all plant fragments are opaque, indicated by light-absorption values of 91 × 10^{-3} microampere and higher. It is interesting to note that a shift in the light

absorption pattern of plant fragments occur between high-volatile B bituminous coal and high-volatile A bituminous coal. The light-absorption values of plant fragments in the high-volatile B bituminous coal range from 60 to 93 × 10^{-3} microampere, whereas in the high-volatile A bituminous coal the range has been narrowed down to 85 to 93 × 10^{-3} microampere.

The carbonization of the psilate trilete spores is hardly noticeable in the range lignite to high-volatile B bituminous coal. The carbonization rate increases in the range high-volatile B bituminous coal to low-volatile bituminous coal. At and beyond the low-volatile bituminous coal rank pollen-grains and spores become totally opaque.

2. SAND-SHALE SAMPLES

The subsurface carbonization trend of a specific psilate trilete spore has been recorded in two sections perpendicular to the strike in the Texas Gulf Coast (figures 17 and 18). In the first section (figure 17) south of Houston, Texas, cuttings samples of eight wells have been analyzed, while in the second section thirteen wells have been analyzed. At specific depths of each well light-absorption measurements of specimens of a psilate trilete spore were made.

The results of these measurements have been plotted on data sheets (figure 19 and 20). For each sample analyzed the carbonization mean and the approximate 95 % confidence limit of this mean have been calculated and plotted on the data sheets (figures 19, 20 and 21).

The carbonization means then have been drawn at specific depths in the wells on the sections (figures 17 and 18).

Isocarbonization lines connecting equal carbonization values in the wells were constructed.

Two obvious observations can be made while studying the carbonization pattern in the two Texas Gulf Coast sections. First of all, there is a clear trend of increasing carbonization with depth. Hilt (in Raistrick and Marshall, 1939), after analyzing coals of the Pas de Calais field, northern France, came to the conclusion that in a vertical sequence at any point in the coal field, the rank of the coals increased with depth, a principle commonly known as Hilt's law. Hilt's law seems to be true for the two sections analyzed in the Texas Gulf Coast.

The second observation is that isocarbonization zones cross time lines. Similar observations have been made by Levenshteyn (1962) in the Donets-basin (figure 22) Russia, and by M. and R. Teichmüller (1958) in the "Bochumer Groszmulde des Ruhrkarbons" east of Bochum, German Federal Republic.

In the Texas Gulf Coast the isocarbonization lines dip
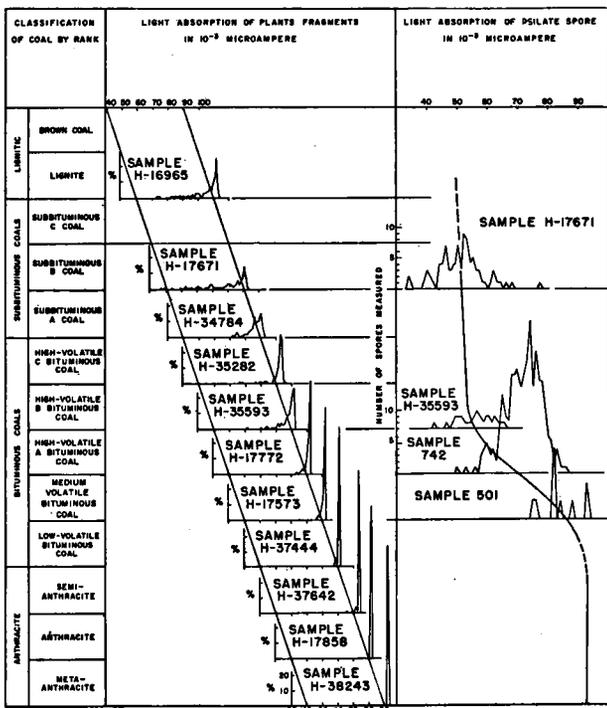
Fig. 15. Light-absorption measurements of plant fragments and psilate spores in humic coals of different ranks.

	Sample	Mine	State	County	Country	Strata
Lignite	H-16965	Kincaid	North Dakota	Barke	U.S.A.	Fort Union
Sub-bituminous B coal	H-17671	Barnham No. 3 coal crop fire project	New Mexico	San Juan	U.S.A.	Fort Union, Mesa Verde
Sub-bituminous A coal	H-34784	Pittsburg and Midway	Colorado	Routt	U.S.A.	Mesa Verde
High-volatile C bituminous coal	H-35282	Orient No. 2	Illinois	Franklin	U.S.A.	Carbondale
High-volatile B bituminous coal	H-35593	Caney Creek	Kentucky	Muhlenberg	U.S.A.	Carbondale
High-volatile A bituminous coal	H-17772	C.C. Conley	West Virginia	Nicholas	U.S.A.	Coalburg in Kanawha group Pottsville Series
Medium-volatile bituminous coal	H 17573	Brubaker	Pennsylvania	Cambria	U.S.A.	Lower Kittanning in Allegheny formation
Low-volatile bituminous coal	H 37444	Pageton	West Virginia	McDowell	U.S.A.	Pottsville
Semi-anthracite	H37642	Glen Burn	Pennsylvania	North Humberland	U.S.A.	Pottsville
Anthracite	H 17858	Greenwood Breaker	Pennsylvania	Schuylhill	U.S.A.	Pottsville
Meta-anthracite	H38243	Granston	Rhode Island	Providence	U.S.A.	Kingston

	Sample	Town	Area	Country	Strata
	742	Wellendorf	Teutoburger Wald	German Federal Republic	Wealden
	501	Rainsen	Wesergebirge	German Federal Republic	Wealden

Fig. 14. Coal data.

Age	Moisture (as received)	Volatile matter (moisture free)	Fixed carbon (moisture free)	Ash (moisture free)	Sulphur (moisture free)	British thermal units (moisture free)
Paleocene	18.2%	42.0%	42.9%	15.1%	1.3%	10560
Cretaceous	8.5%	39.2%	41.9%	18.9%	0.6%	8700
Cretaceous	7.8%	40.9%	49.4%	9.7%	0.6%	
Pennsylvanian	5.4%	36.2%	55.9%	7.9%	1.5%	13110
Pennsylvanian	5.2%	44.2%	49.1%	6.7%	3.5%	13510
Pennsylvanian	1.8%	37.4%	55.6%	7.0%	0.6%	14160
Pennsylvanian	0.6%	21.3%	70.8%	7.9%	1.6%	14430
Pennsylvanian	0.8%	20.8%	74.6%	4.6%		14930
Pennsylvanian	2.2%	8.3%	81.0%	10.7%	0.8%	13480
Pennsylvanian	2.4%	3.4%	88.0%	8.6%	0.5%	13550
Carboniferous	0.3%	2.7%	59.5%	37.4%	0.2%	8510

Age	Volatile matter (in vitrite)	Carbon (in vitrite)	Oxygen (in vitrite)	Hydrogen (in vitrite)	
Cretaceous	31.0%	88.0%	5.8%	6.2%	
Cretaceous	22.0%	92.0%	2.4%	5.6%	

Fig. 14. Coal data (continued).

Fig. 16.　Well locations and index.

SECTION 1

1. Davis and Co. Inc. No. 1, John R. McLane
2. Shell No. 2, C. R. Schiurring
3. Shell No. 1, H. M. Kane
4. Shell No. 1, C. W. McDermott, et al.
5. Colorado No. 1, Pryor, et al.
6. Texas No. A-33, Pierce Est.
7. Pure No. 1, Mrs. E. Page
8. Sun No. 1, Pierce Est.
9. Union No. 1, Pinkney
10. Old Ocean No. 1, S. C. Patterson
11. Pan American Old Ocean Unit No. 99
12. Pan American Old Ocean Unit No. 138
13. Pan American Old Ocean Unit No. 77

SECTION 2

14. Gulf No. 1, Katie Ward
15. Pan American No. 1, Sneed
16. Hammond No. 1, Tieman
17. Pan American No. 1, Suderman
18. Phillips No. 2 M, Houston Farms
19. Phillips No. 1 L, Houston Farms
20. Phillips No. 1 Y, Houston Farms
21. Sun No. 1, Houston Farms

Fig. 17.　Variations in the carbonization degree of psilate spores in Section 1, Texas Gulf Coast, U.S.A.
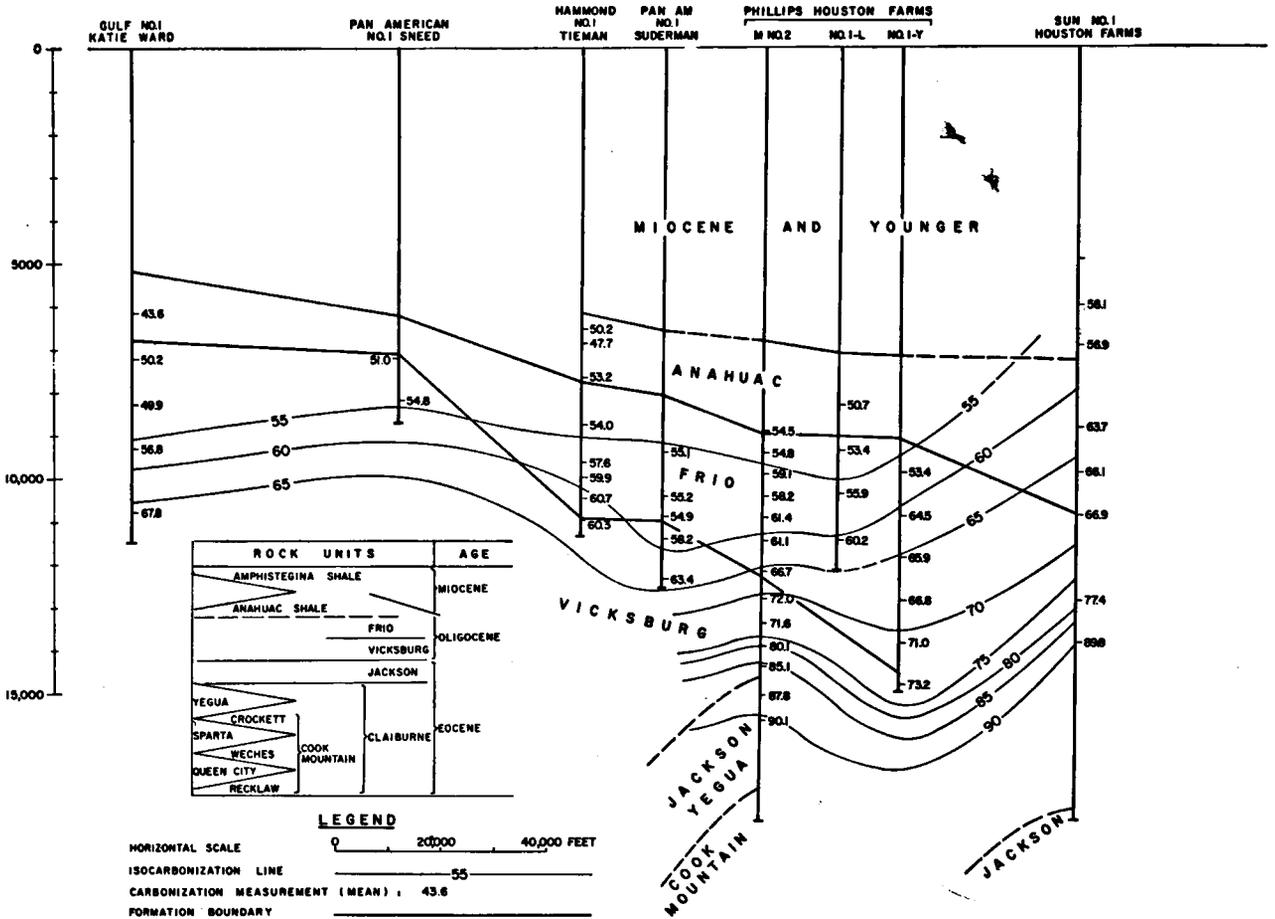
Fig. 18. Variations in the carbonization degree of psilate spores in Section 2, Texas Gulf Coast, U.S.A.

less towards the coast than the time lines. Therefore, rock units like Wilcox, Yegua, Vicksburg, Frio, etc., are less carbonized in their updip areas, and consequently are more carbonized in their downdip directions. If the carbon ratio theory is being applied in the Texas Gulf Coast, oil pools may disappear in the downdip direction of each formation.

Examples (figure 1) showing the relation of commercial oil fields and the fixed carbon percentage of coals, indicate the disappearance of big commercial oil pools on a world-wide basis in sediments containing coal with a fixed carbon of 65—70 and higher values. The 70 % fixed carbon can be correlated approximately with the 72×10^{-3} microampere light absorption [1] of a specific trilete psilate spore (figure 15).

Using above correlation, it is interesting to observe that the Frio Formation in Section 2 (figure 18b) enters the 65—70 fixed carbon values (estimated

equivalent of $67—72 \times 10^{3}$ microampere light absorption value) in the area Phillips Houston Farms M No. 2 and Sun No. 1, Houston Farms. The Vicksburg Formation reaches similar values updip of the Phillips Houston Farms wells. The Jackson, Yegua, and Cook Mountains Formations in Section 2 (figure 18) are severely carbonized varying from 86×10^{-3} microamperes light absorption to totally opaque. The Miocene and younger, and Anahuac have not reached the values 67 to 72×10^{-3} microampere light absorption in this section (figure 18).

In Section 1 it appears that the Eocene rock units (Carrizo-Wilcox, Cook Mountain, and Yegua) are dipping into the 67 to 72×10^{-3} microamperes light-microamperes light-absorption values updip of well Pure, No. 1 Mrs. E. Page. The Oligocene units (Vicksburg and Frio) will encounter similar values somewhere between the wells Texas, No. A33, Pierce Est and Pure, No. 1, Mrs. E. Page and in the downdip direction of these wells (figure 17). The Miocene and younger strata in this section are lightly carbonized (less than 55×10^{-3} microampere).

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1 It must be emphasized that those light-absorption values were obtained with a particular microscope equipped for carbonization studies. If other microscopes are used, calibration is a necessity if results are being compared.

WELL : DAVIS AND CO. INC. NO. I J. R. MC LANE C = CORE
CUT = CUTTINGS
COLORADO COUNTY, TEXAS

SAMPLE DEPTH	LIGHT ABSORPTION IN 10⁻³ MICROAMPERES	95% CONFID. LIMITS OF MEAN	AVERAGE MEASUREMENT
Cut 4512-4540		48.10-52.50	49.30
" 5019-5049		47.10-53.30	50.27
" 5327-5359		53.20-59.40	56.36
" 5514-5544		49.60-54.40	52.05
" 6314-6349		58.10-64.70	61.46
" 6504-6534		57.50-62.70	60.13
" 7028-7058		57.60-63.60	60.66
" 7275-7305		56.20-64.80	60.52
" 7523-7553		60.00-65.60	62.84
" 8020-8050		61.80-64.70	62.78
" 8514-8544		65.40-71.20	68.36
" 9527-9535		65.70-74.70	70.25
" 10,008-10,026	Poor		
" 10,465-10,481	Barren		

WELL : SHELL OIL CO. NO. 2 SCHIURRING
COLORADO COUNTY, TEXAS

SAMPLE DEPTH		95% CONFID.	AVERAGE
Cut 2999-3029		44.60-53.70	49.00
" 4001-4033	Barren		
" 4517-4548			
" 5044-5095		51.70-55.80	53.77
" 5469-5499		45.50-55.30	50.43
" 5995-6057		58.00-62.80	60.45
" 6556-656?		51.00-57.80	54.43
" 7021-7083		60.40-65.20	62.85
" 7519-7550		56.80-65.10	60.94
" 7985-8047		62.80-68.00	65.40
" 8514-8545		58.10-67.70	62.92
" 8978-907?			
" 9521-9552			
" 9919-9981		65.90-73.70	69.45

WELL : SHELL OIL CO. NO. I H. M. KANE
COLORADO COUNTY, TEXAS

SAMPLE DEPTH		95% CONFID.	AVERAGE
Cut 4017-4048		45.30-49.90	47.65
" 4999-5060		51.10-54.70	52.95
" 5522-5553		50.50-55.10	52.86
" 6014-6076		49.20-58.70	53.02
" 6504-6535		46.10-54.00	50.06
" 7027-7077		55.20-60.50	57.92
" 7507-7528		55.70-64.70	60.26
" 8001-8061		54.30-62.10	58.23
" 8523-8553		60.00-61.80	58.88
" 8985-9046		60.50-70.50	65.56
" 10,000-10,031		64.40-70.80	67.61
" 10,523-10,553	Barren		
" 10,903-10,9??	Barren		

WELL : OIL DRLG. (SHELL) NO. 2 C. W. MC DERMOTT ET AL
COLORADO COUNTY, TEXAS

SAMPLE DEPTH		95% CONFID.	AVERAGE
Cut 4477-4507		43.20-64.80	54.00
" 5026-5056		52.10-57.50	54.84
" 5520-5550		50.80-56.00	53.45
" 6009-6070		58.50-61.30	58.91
" 6529-6559		48.90-59.90	54.40
" 6975-7046		55.50-60.50	58.10
" 8024-8085		56.60-63.60	60.15
" 8510-8544		62.90-66.40	64.68
" 9035-9096		49.40-75.40	62.42
" 10,142-10,193		63.90-69.30	66.61
		65.20-74.20	69.72

WELL : FEDERAL ROYALTY AND A. WELCH CO. NO. I MYRIA PRIOR
WHARTON COUNTY, TEXAS

SAMPLE DEPTH		95% CONFID.	AVERAGE
Cut 5420-5573		47.30-52.10	49.71
" 5995-6026		50.40-55.20	52.84
" 6475-6505		52.50-57.30	54.91
" 7091-7121		53.90-60.30	57.10
" 7514-7576		56.40-60.30	58.40
" 8481-8510		59.50-64.90	62.20

WELL : TEXAS CO. NO. A-33 PIERCE EST.
WHARTON COUNTY, TEXAS

SAMPLE DEPTH		95% CONFID.	AVERAGE
Cut 5574-5578		40.20-43.80	41.90
" 6065-6097		39.10-59.10	49.14
" 6487-6518		43.20-57.60	50.44
" 6826-6857		48.70-53.90	51.36
" 7305-7315		50.80-56.80	53.70
" 7895-7899		55.70-59.10	57.44
" 8314-8324		54.90-61.30	58.11
" 8853-8860	Poor		

Fig. 19. Light-absorption data of psilate spores from wells in Section 1.

WELL : PURE OIL CO. NO. I MRS. E. PAGE
WHARTON COUNTY, TEXAS

C = CORE
CUT = CUTTINGS

SAMPLE DEPTH	LIGHT ABSORPTION IN 10^{-3} MICROAMPERES	95% CONFID. LIMITS OF MEAN	AVERAGE MEASUREMENT
Cut 3828-3846	Barren		
" 4515-4535	Barren		
" 5047-5067	Barren		
" 5492-5494		52.10-58.10	55.19
" 6329-6399	Poor		
" 6359-6379	Barren		
" 7220-7360		67.40-72.00	69.74
" 7873-7883		51.80-63.60	57.71
" 8280-8340		56.00-62.20	59.18
" 8820-8830		56.40-62.00	59.25
" 9170-9190		67.10-70.90	69.04

WELL : SUN OIL CO. NO. I PIERCE EST.
MATAGORDA COUNTY, TEXAS

SAMPLE DEPTH		95% CONFID. LIMITS	AVERAGE
Cut 5283-5314		42.70-55.10	48.90
" 6373-6403		46.90-55.90	51.43
" 7427-7458		50.70-56.90	53.83
" 7953-7984		52.00-57.00	54.52
" 8387-8417		49.90-57.50	53.74
" 8975-9005		58.50-65.90	62.26

WELL : UNION PROD. NO. I PINKNEY
MATAGORDA COUNTY, TEXAS

SAMPLE DEPTH		95% CONFID. LIMITS	AVERAGE
Cut 8085-8115		48.20-55.00	50.66
" 8515-8546		49.20-55.20	52.20
" 9002-9034		52.40-57.00	54.74
" 9502-9512		50.00-61.00	55.57
" 9918-10044		52.70-57.50	55.13
" 10,502-10,532		50.80-68.00	59.30
" 10,759-10,990		51.80-57.80	54.73

WELL : OLD OCEAN DEV. CO. NO. I S.C. PATTERSON
BRAZORIA COUNTY, TEXAS

SAMPLE DEPTH		95% CONFID. LIMITS	AVERAGE
Cut 6086-6116		47.40-50.70	49.13
" 6508-6538	Poor		
" 7032-7062	Poor		
" 7522-7552		36.30-59.30	47.86
" 8014-8075		44.40-55.20	49.82
" 8505-8536	Poor		
" 9059-9121		51.70-57.90	54.68
" 9521-9551	Poor		
" 10,036-10,067			
" 10,339-10,370		59.10-65.70	62.80

WELL : PAN AMERICAN NO. 99 OLD OCEAN
BRAZORIA COUNTY, TEXAS

SAMPLE DEPTH		95% CONFID. LIMITS	AVERAGE
Cut 5966-5999		40.50-56.10	48.37
" 6909-6939		44.60-51.20	47.94
" 7974-8004	Poor		
" 8975-9005		45.10-52.90	49.09
" 9993-10,024		42.10-57.90	55.09
" 10,942-10,957		53.60-71.20	67.44

WELL : PAN AMERICAN NO. 138 OLD OCEAN
BRAZORIA COUNTY, TEXAS

SAMPLE DEPTH		95% CONFID. LIMITS	AVERAGE
Cut 8329-8360		48.20-63.70	56.00
" 9375-9405		49.80-57.80	53.80
C 10,405-10,957		59.10-72.10	65.87

WELL : PAN AMERICAN CO. NO. 77 OLD OCEAN UNIT
BRAZORIA COUNTY, TEXAS

SAMPLE DEPTH		95% CONFID. LIMITS	AVERAGE
Cut 7003-7061		41.90-55.70	48.87
" 7509-7539		42.40-52.40	47.44
" 8022-8082	Poor		
" 8503-8533		44.80-53.80	49.18
" 9019-9139		50.30-56.50	53.42
" 9529-9559	Poor		
" 10,007-10,037		53.80-60.00	56.95
" 10,189-10,219		50.00-66.40	58.28
" 10,518-10,548		47.90-64.30	56.14
" 11,000-11,030	Poor		
" 11,500-11,530		54.60-58.40	56.50
" 12,040-12,070	Barren		
" 12,520-12,590		58.10-61.00	59.61
" 13,000-13,030			
" 13,480-13,510			
" 14,020-14,140			
" 14,500-14,530			
" 15,010-15,040		74.90-84.30	79.62

Fig. 20. Light-absorption data of psilate spores from wells in Section 1.

WELL : GULF OIL CO. NO. I KATIE WARD
HARRIS COUNTY, TEXAS

C = CORE
CUT = CUTTINGS

SAMPLE DEPTH	LIGHT ABSORPTION IN 10^{-3} MICROAMPERES	95% CONFID. LIMITS OF MEAN	AVERAGE MEASURE-MENT
Cut 6172-6202		39.62-47.62	43.62
" 7249-7280		47.46-53.06	50.26
" 8340-8370		47.13-52.73	49.93
" 9356-9385		53.79-59.99	56.89
" 10,871-10,904		64.25-71.25	67.85

WELL : PAN AMERICAN CO. NO. 4 SNEED
BRAZORIA, TEXAS

SAMPLE DEPTH		95% CONFID.	AVERAGE
Cut 5130-5190	Poor		
" 6153-6190	Poor		
" 7225-7255		48.04-54.04	51.04
" 8250-8280		51.46-58.26	54.86

WELL : HAMMOND NO. I TIEMAN
BRAZORIA COUNTY, TEXAS

SAMPLE DEPTH		95% CONFID.	AVERAGE
Cut 6479-6508		46.15-54.35	50.25
" 6907-6937		44.18-51.38	47.78
" 7740-7770		48.95-57.55	53.25
" 8258-8288	Poor		
" 8807-8837		52.08-55.98	54.03
" 9727-9751		55.62-59.72	57.67
" 10,060-10,090		57.68-62.28	59.98
" 10,555-10,583		58.05-63.45	60.75
" 11,015-11,046		58.07-62.67	60.37

WELL : PAN AMERICAN NO. I SUDERMAN
BRAZORIA COUNTY, TEXAS

SAMPLE DEPTH		95% CONFID.	AVERAGE
Cut 8990-9020			
" 9470-9500		52.05-58.15	55.10
" 10,490-10,530		52.37-58.17	55.27
" 10,970-11,000		49.41-60.41	54.91
" 11,480-11,510		53.59-60.99	58.29
" 12,440-12,470		61.37-65.57	63.47

WELL : PHILLIPS M-NO. 2 HOUSTON
BRAZORIA COUNTY, TEXAS

SAMPLE DEPTH		95% CONFID.	AVERAGE
Cut 8979-9040		52.08-57.08	54.58
" 9474-9536		50.19-59.59	54.89
" 9990-10,040		56.83-61.43	59.13
" 10,524-10,554		54.95-61.55	58.25
" 10,982-11,073		59.54-63.44	61.49
" 11,560-11,620		57.11-65.11	61.11
" 12,260-12,320		62.89-70.69	66.79
" 12,990-13,080		70.33-73.83	72.08
" 13,480-13,540		69.64-73.64	71.64
" 13,990-14,050		76.99-83.39	80.19
" 14,500-14,560		83.17-87.07	85.12
" 15,180-15,240		84.80-90.80	87.80
" 15,780-15,810		87.93-92.93	90.18
" 17,020-17,050	Barren		
" 17,940-17,970	Barren		

WELL : PHILLIPS NO. I-L HOUSTON FARMS
BRAZORIA COUNTY, TEXAS

SAMPLE DEPTH		95% CONFID.	AVERAGE
Cut 8387-8417		42.05-59.45	50.75
" 9460-9490		49.68-57.18	53.43
" 10,472-10,502		53.26-58.66	55.96
" 11,560-11,589		58.15-62.25	60.20

WELL : PHILLIPS NO. I-Y HOUSTON FARMS
BRAZORIA COUNTY, TEXAS

SAMPLE DEPTH		95% CONFID.	AVERAGE
Cut 9970-10,000		50.31-56.51	53.41
" 10,980-11,010		60.72-68.32	64.52
" 11,980-12,010		60.92-70.92	65.92
" 12,970-13,000		63.90-69.70	66.80
" 13,990-14,020		67.79-74.39	71.09
" 14,950-14,990		70.60-75.80	73.20

WELL : SUN OIL CO. NO. I-HOUSTON FARMS
BRAZORIA COUNTY, TEXAS

SAMPLE DEPTH		95% CONFID.	AVERAGE
Cut 6062-6093		50.93-61.43	56.18
" 7021-7052		49.68-64.18	56.93
" 8960-8992		58.20-69.27	63.70
" 10,025-10,057		63.54-68.74	66.14
" 11,028-11,059		60.17-73.67	66.92
" 13,024-13,054		70.91-83.91	77.41
" 14,007-14,039		87.05-92.55	89.80
" 15,013-15,043	Barren		
" 16,015-16,044	Barren		
" 17,024-17,053	Barren		
" 17,960-17,990	Barren		

Fig. 21.   Light-absorption data of psilate spores from wells in Section 2.
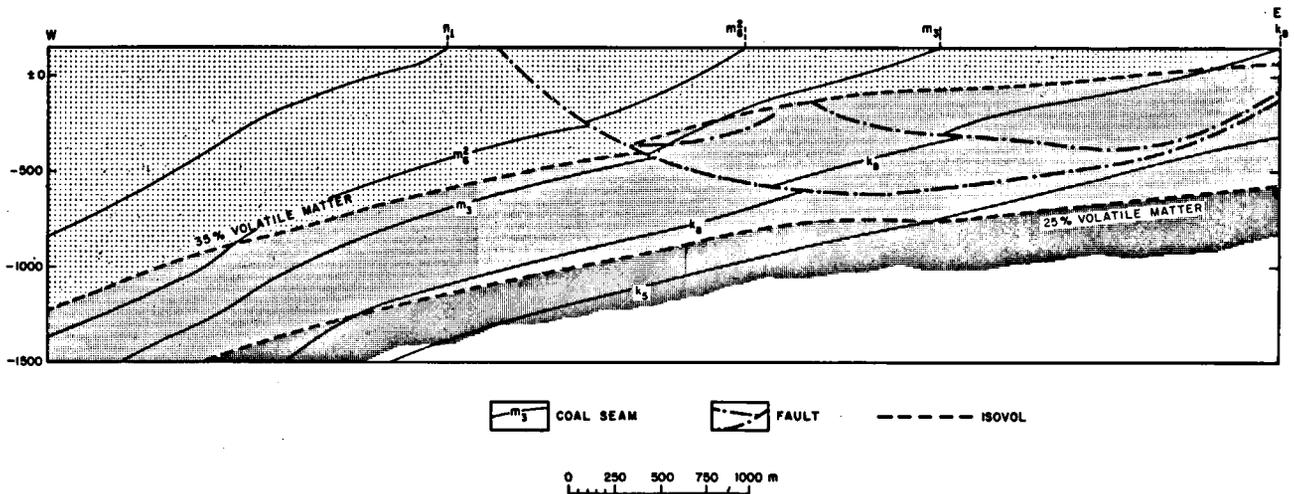
Fig. 22. Variations in degree of metamorphism of coals in the central region of the Donets basin (west of Main anticline,) Russia. (Levenshteyn 1962).

Whether 67 to 72 × 10^{-3} microampere light-absorption values can be used in the Texas Gulf Coast to indicate a transition zone between big commercial oil accumulations and small non-commercial oil accumulations or no oil at all (but condensates and gas only or nothing a all) will be tested by deeper drilling in future, which will provide the additional data necessary to confirm or disprove the above-mentioned values.

As more data become available in the future, it may be necessary either to raise or lower the values indicated above. The data so far used are based upon the relation between fixed carbon of coals (and their equivalents expressed in the light absorption of psilate, trilete spores) and the occurrence of commercial and noncommercial or absence of oil pools on a world-wide basis.

CHAPTER VII

APPLICATIONS

1. CORRELATION BETWEEN CARBONIZATION AND OTHER SEDIMENT PROPERTIES

In addition to the correlation between the carbon ratios of coals and the carbonization of pollen-grains, spores, and other plant material, the latter might exhibit a number of other interesting correlations with sediment properties.

A correlation between porosity and carbonization has been noticed by several authors. Weber (in Koslow, 1958) reports a relationship between sandstone porosities and carbon ratios of associated coals in the northern part of the Donets basin of Russia (figure 23). However, no correlation between sandstone porosity and volatile matter of coals in the Ruhr area (West Germany) has been reported by R. Teich-müller (1962).

Russel (1951) reports a correlation between carbon ratio and porosity in sandstone which shows that for loose, porous and permeable sandstone, carbon ratios of associated coals generally fall below 60. For values between 60 and 80, the sandstones are usually very

hard and show only local porosity. Very hard quartzitic sandstones and quartzites occur with coals having carbon ratios greater than 80. It should also be recognized, however, that low-porosity sediments can be found in rocks with a low-carbonization value caused by the clay content of the sands, early cement-tation, etc.

Peldjakow (in Koslow, 1958) has reported average specific gravities of 2.17-2.35 for rocks occurring in sequences with brown coal and medium volatile bituminous coal in the Donets and Kuznetsk basins and in the Ural region. Characteristically, the average specific gravities of the rocks in the above-mentioned areas, intercalated with higher rank low-volatile bituminous coals range betwee 2.35 and 2.60. The average specific gravities of sandstones associated with high-rank semi-anthracites and anthracites are 2.60—2.75 in the Donetx and Kuznetsk basins and in the Ural region.

Koslow (1958) reports that sicsmic velocities of dediments occurring with brown coal and lower rank
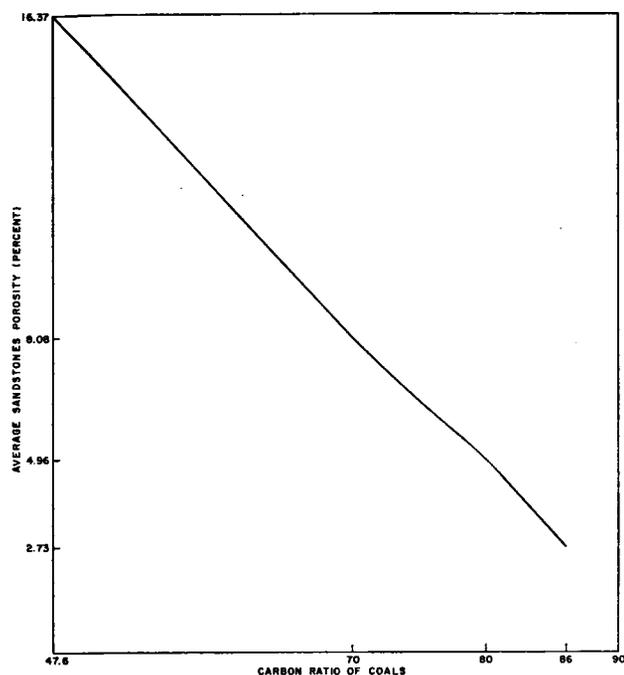
Fig. 23. Relationship between sandstone porosities and carbon ratios of coals in the northern part of the Donets basin, Russia. (Koslow 1958).

semi-anthracites vary from 2600 to 3550 meters per second. Dediments containing high-rank semi-anthracites to anthracites show seismic velocities of 4000 to 5000 meters per second. However, high-pressure shale zones may show moderately to strongly carbonized pollen-grains and spores but have lower seismic velocities and better sandstone porosities than expected for the present depth of burial. We emphasize that low porosities of sandstones, high specific gravities, high seismic velocities of the sediments, etc., can also be due to other factors which do not cause carbonization of pollen-grains and spores. Therefore, sediments with these properties do not invariably have highly carbonized pollen-grains and spores. Furthermore, intensely carbonized pollen-grains and spores may occur in sediments with secondary porosity.

2. PALYNOLOGICAL APPLICATION

A reasonable to optimum preservation of pollen-grains and spores is a necessity for palynological correlations and ecological studies. Strongly to intensely carbonized pollen-grains and spores are opaque and show no structure. They are, therefore, difficult to recognize and palynological correlations and ecological investigations in areas in which they occur are difficult and often impossible. Because severe carbonization is a limiting factor in carrying out palynological correlation or ecological studies, it is obvious that the delineation of severely carbonized areas is important to the palynologist and to the exploration effort in general. In basins in which no palynological studies have been carried out, the rank of the coals might help the palynologist to select favorable areas for palynological standard sections. Samples collected for the purpose of palynological time-correlation or ecological studies ought not to be taken from sediments which level of organic metamorphism is equivalent to semi-anthracite, anthracite or meta-anthracite. Samples taken from sediments which level of organic metamorphism is equivalent to medium-volatile coal and low-volatile coal may contain brown to dark brown spores or pollen-grains. None to slightly carbonized pollen-grains and spores can be expected in sediments with levels of organic metamorphism equivalent to the range lignite to high-volatile A bituminous coal.

3. OIL INDUSTRY APPLICATION

This study suggests that the degree of carbonization of pollen-grains and spores in sedimentary rocks devoid of coals can be used instead of fixed carbon values to determine areas of equal organic metamorphic intensity and to provide a clue to the hydrocarbon potential of these areas. If important commercial oil accumulations are found in slightly to not metamorphosed sediments, and if other severely metamorphosed sediments of essentially the same character are devoid of important oil accumulations, metamorphism may have converted the sediments from a productive to an unproductive condition (Thom, 1934).

Pollen-grains and spores can be used as recorders of incipient rock metamorphism and, as such, can delineate intervals and areas within which certain metamorphic intensities have been reached. Although sharp and definite boundaries to economical oil accumulations do not exist, it is probable that significant economical oil occurrences will decrease progressively as intensity of organic metamorphic alteration increases (Thom, 1934).

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