

THE CONSTITUTION OF COAL

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In regard to the knowledge of the constitutions of coal we are still in the Dark Ages. To the man in the street, coal is merely a dirty mass of combustible matter; to the engineer it is generally thought of in terms of its contents in carbon, volatile matter, moisture, ash and sulphur; and to the geologist, it is merely regarded as a stratigraphical unit. But what is coal? What distinctive compounds or constituents can be proved to exist as such in the coal mass? Investigators of such problem during the past decades have no doubt done much of the pioneer work, crude and contradictory sometimes though it may be; but it is due to the work of recent investigators that we can say we now get an inkling of the real constituents that make up the coal mass. Among many others may be mentioned the following prominent research workers in this field: in the United States, White, Thiessen Porter, Parr; in England, Stopes, Wheeler, Lessing, Illingworth, Lomax; and in Germany, Fischer, Hofmann, Schrader. Prominent among the organizations which have made a vigorous research into the nature of coal, are the Bureau of Mines in the United States, the Department of Scientific and Industrial Research and the Lancashire and Cheshire Coal Research Association in England, and the Kaiser-Wilhelm-Institut für Kohlenforschung in Mülheim and the Friedländer-Füldches Kohlenforschungsinstitut in Germany. If I am allowed to generalize, I am inclined to think that the tendency of the American research is toward the elucidation of the structure of coal, the English research work leaves rather toward its fragmentary significance and the Germans have done much in the investigation of the chemical decomposition of coal.

On a whole these researches can be grouped into:

1. Megascopic
2. Microscopic {
 - A. Transmitted light
 - B. Reflected light
3. Chemical {
 - A. Chemical attack by means of various reagents.
 - B. Action of solvents
 - C. Low temperature distillation.

4. Photographic

5. X-ray

I. MEGASCOPIC.

The megascopic examination of coal should be, as it is frequently done, supplemented by its microscopic examination. Thanks to the indefatigable labor of Dr. Marie C. Stopes who has done much to classify our conception of the constituents of coal by a broad generalization of separating coal into four ingredients—fusain, durain, vitrain and clarain. Indeed, previous investigators had recognized some of these ingredients but their descriptions were oftentimes rather loose and vague. Before going into the details of these four ingredients I shall first try to correlate the nomenclatures as adopted by the few who have studied this problem.

Stopes:	<i>Fusain</i>	<i>Durain</i>	<i>Vitrain</i>	<i>Clarain</i>
Hutton:	dull coal	dull coal	crystalline coal	crystalline coal
Dawson:	Mineral charcoal	compact coal		
Haxley:	Mineral charcoal	duller slate coal	bright conchoidal pitch	
		coal proper		
Wetherell:	dull coal		bright coal	
	— microspores & megaspores		— hydrocarbon	
Potonie:	dull or mat coal		humus coal (glantz or bright coal)	
Thiessen:		dull coal & attritus	anthroxylon	
Lomax:			jetonised wood	

Fusain is a jet-black, opaque, flocculent porous powder, somewhat fibrous, disseminating in patches or wedges along the bedding plane of coal, from a thin layer to bands several inches in thickness. In contradistinction to it, bituminous coal, when ground to extreme fineness, is of a dark mahogany color and its particles are more or less rounded. Sometimes fusain occurs "as an aggregate of material" not easily to be pulverized. It is a product of the ridding of "the hard portions of plants, such as the liquified cell walls to

moleucules approximating carbon" on the surface of water where the oxygen of the air had free play.

Durain is a dull-appearing coal substance, having a hard, close and firm texture appearing rather granular to the naked eye. It breaks irregularly across the bedding plane with finely lumpy surface. Durain bands are variable in thickness, occasionally occurring in long lenticular shape. In some cases it may be confused with clarain especially when numerous thin bands of vitrain are intermixed with the dull coal substance. However, vitrain may be recognized as such by having the coal fractured along the bedding plane. In fact durain represents the ultimate residues of the disintegrated plants, in which "the potentially vitrain forming portion" mingles freely.

Vitrain: In the words of Stopes: "Just the essential point of difference between vitrain and ordinary bright coal (which I have called clarain) lies in the fact that the vitrain bands, unlike clarain, are entirely devoid of preserved structure. Vitrain is indeed a solidified jelly, and is practically homogeneous under the highest power of microscope. That is, it is, at any rate, so far jellified that, whatever plant material had originally composed it, no vestige of that cellular structure is still extant. Vitrain occurs as definite, rather narrow bands in some instances straighter and flatter than the other bands of coal, and in some instances more obviously lenticular. True brilliant vitrain bands are often markedly uniform in thickness for considerable distances." Vitrain is then a colloid-like substance, that can be broken into more or less regular cubes, representing the product of bacterial and chemical disintegration of plants, in the absence of oxygenated water or free atmospheric oxygen, but with sufficient moisture.

Clarain: The characteristic of clarain is its streakiness, or contrasted with the brilliant and structureless character of vitrain. In the words of Stopes, "clarain possesses a definite and smooth surface when broken at right angles to the bedding plane and these surfaces have a pronounced gloss or shine. This surface luster is seen to be inherently bonded as well as to have bands of fine durain intercalated between its own bands". It is supposed that durain was formed in the central portions of wide, still lagoons in which the plants were more or less completely decayed while materials such as cuticles, spore-exines and carbon that were more resistant to the decaying

process, mingled as such with the more delicate tissues.

If these four ingredients now recognized so by many observers, are really distinctive substances (they are mere substances in the true sense of the word because each one of them may be eventually resolved into some definite chemical entities) they should possess, besides their apparent structural characteristics, both chemical and physical properties to distinguish one from another. Indeed, they possess such distinguishing properties. Lessing found in one of the samples of bituminous coal examined, the following percentages of ash, which may be taken as indicating the relative ash contents of the four ingredients:

Fusain	Durain	Clarain	Vitrain
15.59	6.26	1.22	1.11

In a general way, coal dust is made up mostly of fusain, the lighter coal is composed of a combination of vitrain and clarain and the heavier coal is represented by durain. Lessing stated, "the ash constituted by the dust was at least equal to and in some cases more than twice as much as that constituted by the whole bulk of the clean coal". Tideswell and Wheeler determined their volatile matter in percentages as follows:

Fusain	Durain	Clarain	Vitrain
22.6	39.4	40.8	36.6

Sinvatt drew the following conclusion from a study of the numerous analyses of the four ingredients. Fusain has the highest ash and lowest volatile matter; durain has lower moisture and higher ash than either vitrain and clarain; clarain has higher and sometimes lower volatile matter and ash and lower moisture than vitrain; vitrain has the highest moisture and lowest ash.

According to Sinvatt the agglutinating values of the Aseley bituminous coal are:

Coal	Vitrain	Clarain	Durain	Fusain
15	9	17	6	nil

that is to say, fusain is absolutely non-coking, durain is slightly coking with particles adhering slightly together, clarain fuses considerably with the merging together of particles giving rise to the peculiar brown shear to cokes, and vitrain cokes and is responsible for the silver-white appearance of cokes.

Regarding the relative abundance of the four ingredients in a seam of coal, mention should be made of the Wigan Four Feet Seam, which according to Sinvatt gives the following percentages: Vitrain 25, Clarain 52, Durain 20, Fusain 1. However, it should be emphasized, such percentages are always very variable with different seams or with different parts of the same seam. Nevertheless I summarise that, whatever variabilities there are in the percentages and arrangements of the four ingredients in a seam of coal, there might be established, as we advance further in the regional observations of these ingredients, an index from their percentages or some kind of criteria from their arrangement, by means of which one seam can be correlated with another.

As to the occurrence of these ingredients in anthracite, we are indebted to Grounds for his observations which I now summarize below: Anthracite contains some fusain, although not in such great quantity as is found in bituminous coal. It has been pointed out that the volatile matter of the fusain in bituminous coal is generally lower than that of its associated coal. The clarain is very similar to that of bituminous. Grounds made out another new constituent, which he called Ψ vitrain or pseudo-vitrain. Examined under the microscope, it appears deep brown and much darker than the vitrain of bituminous. It breaks with a conchoidal fracture into sharp splinters and fragments of pyramidal shape, whereas vitrain proper breaks more or less regularly into cubes; it is much harder than vitrain proper. If the fusain is found to contain higher percentage of ash than the coal, then its volatile matter is higher than that of Ψ vitrain; and, *pari passu*, if lower, then latter. Ψ vitrain forms the major portion of anthracite while fusain 1% or less and clarain 5—10%.

II. A. MICROSCOPIC EXAMINATION OF COAL BY MEANS OF TRANSMITTED LIGHT.

Fusain: black, opaque, showing little cellular structure of wood; walls thickened and cell lumina empty.

Durain: granular matrix of roundish or polyhedral fragments, mixed with spore exines; macrospores conspicuous, whose exines are clear and brilliantly colored, almost red; small amount of microspores; sometimes

clarain presents as clear lenticular bands or flecks of golden color; a little slightly colored bodies, supposed to be resin.

Clarain: clear, as contrasted with opacity of durain; bands of disintegrated plant substance and bands of clear cuticle spore exines, resin bodies and other structures of various shades, from pale yellow to a rich amber-reddish, stem tissues and leaf tissues; durain streaks presents as impurity.

Vitrain: translucent, structureless, uniform texture; color ranging from pale yellow to ruddy brown, depending upon thickness of slide.

Let us now examine the structures of the different elements of plants which make up the coal mass.

Cuticle: "Morphologically cuticles are very different from cork tissue, they retain in ridges and sculpturing the impress of each individual cell it overlays". Hence a sheet of cuticle looks like a sheet of cells. A perpendicular section would show the cuticle to be saw-edged on one side and smooth on the other. It is bright yellow or orange in color, clear and translucent, in very thin section it is pale yellow. It is considerably long and relatively narrow. White and Thiessen considered the cuticle to be resinous in composition, while Stopes and Wheeler considered it to be waxy.

Bark may be recognized by its lumpy porous and irregular structure. It is dark brown in color.

Spore-coats or spore-exines have flattened oval outline. They grade from a brilliant yellow to a ruddy orange in color. Although circular or oval in general appearance, they tend to become triangular in shape with tetrasporic marks which distinguish them from pollen-exines whose surfaces are smooth with a long slit parallel to the axis of the ovoid. Spores range from 10 microns ($\frac{1}{100}$ mm) in diameter, i.e., microspores, to 2 and 3 mm., i.e., megaspores. Spores are found in immense quantity in cannel or boghead coals.

Rodlets are circular or oval in cross-section, which is relatively larger than that of spores. They are black, glistening or petrified contents of the mucilage canals of certain plants.

Resines. Microscopically their recognition is exceedingly difficult and uncertain. Concerning resines, White said, "There are distinctly two kinds—one dull, opaque, non-refractive; the other, (may be of a waxy nature)

light and very refractive. Of these the former is by far the more abundant". According to White and Thiessen, resin bodies are generally oval or roundish in cross-section; but on cleavage surfaces they may be seen as rodless, being "resinous fillings of certain true vessels or gum or resin ducts in certain woods". There are in record lumps of resins found in Palaeozoic coals as well as in late aged coals.

Other structural substances such as sporangia (receptacles, in which spores are produced), seed, phloem (soft-walled, proteine-containing cells) all of which, from their very nature, are easily decomposed, and are, therefore, very rarely observed. Other unstable organic substances such as protoplasm, chloroplast, starch grain, oils, sugar etc. may be or most probably may not be present as such in the coal mass and are beyond the power of microscope to render them observable.

Besides the extraneous rocks and minerals such as shale, clay salt etc. forming the impurities found in coal, there are other inorganic products such as carbonates of iron, iron sulphide, calcite, magnesia, sulphur and alumina, which have been extracted from the tissues of the once living plants and deposited in the coal substance, mainly by the action of bacteria and other microorganisms. Of these, sulphur is decidedly the most important and thus warrant a little notice here. According to Prof. Parr, sulphur occurs in coal as pyritic sulphur, in disseminated or microscopic form, organic sulphur, sulphates and possibly also as uncombined sulphur. The microscopic pyritic sulphur varies from 20 to 50 microns in size and appears as cubes. These cubes are undoubtedly secondary, that is, they did not occur as such in the original plants but were deposited there during or after coalification.

The microscopic examination of coal, scientifically valuable and illuminating though it is, should become one of the methods of geologists for the correlation of coal seams. As examples of such work, I may mention the observation of White and Thiessen, which show that the Pittsburg bed contains some characteristic spores which have not been found in any other bed and that the Shelbyville coal is full of megaspores.

II. B. MICROSCOPIC EXAMINATION OF COAL BY MEANS OF REFLECTED LIGHT.

So far known to me, E. P. Reim seems to be the only one who has

made a successful attempt of such work. His method consists of etching or heat-tinting the polished surface of coal, and of examining it by reflected light, just as one does in metallographic work. More should be done to prove the value of such method for the examination of coal.

III. CHEMICAL.

As the chemical investigation of coal concerns the chemists more than the geologists, I shall briefly describe the results of such investigation.

As to the exact nature of the organic compounds composing the coal mass opinions are still, so to say, in a flux. We know that the organic compound found in plants are: carbonhydrates and their derivatives,—the glucosides and tannins, fats and waxes, the essential oils and resins, organic acid and their salts, the proteines, the vegetable bases and alkaloids, and the pigments. But how far such organic compounds have undergone changes due to deoxygenation, polymerisation and dehydrogenation during or after coalification is the work of chemists to find out. They have adopted these modes of investigation:

- a.) Chemical attack by means of various reagents.
- b.) Action of solvents.
- c.) Low temperature distillation.

a.) *Chemical attack by means of various reagents*: Chemists have, in one time or another, made use of sulphuric acid, nitric acid, caustic alkalkies, bromine, ozone and oxygen for such work. The results obtained are meagre and unsatisfactory, because the action of such reagents generally masks up the exact nature of the original constituents.

b.) *Action of solvents*: Important results have been obtained by means of solvents. Burgess and Wheeler have been able to isolate some fundamental constituents from coal, which have been confirmed by many other investigators. I am now going to describe them diagrammatically:

Cellulosic

(Polymerisation and degradation products of original plant cellulose).

N. B. Opposing the above, Fischer and

Resinic.

(Polymerisation and degradation products of original plants resins,

Schrader maintain that lignine degradation products rather than cellulosic substances play an important part in coal constitution (gums and waxes)

<i>Alpha</i> <i>Constituent</i>	<i>Beta</i> <i>Constituent</i>	<i>Gamma</i> <i>Constituent</i>
insoluble in pyridine, forms a large part of the fixed carbon in coke.	soluble in pyridine but insoluble in chloroform, yields a high proportion of volatile matter.	soluble in pyridine and chloroform, readily fusible below 500° C; responsible element in coking, which idea has been attacked by Bone, Pearson etc.

Ulmins, which have been found in peat and lignite, have not been found as such in bituminous coal. In the words of Slopes, "It seems to us probable that during the course of formation of bituminous coals ulmin compounds found during the early stages may undergo not distinction but a series of changes resulting in a progressive increase in carbon-content and that compounds similar in constitution to the ulmins but lacking their characteristic solubility in alkaline solution, exist in bituminous coal."

c.) *Low temperature distillation:* Since the number of the investigators in this kind of research is legion and their results are sometimes complicating, I cannot do better than to condense the summary given by Bone about the present state of our knowledge with regard to the low temperature distillation of coal. There is little doubt about the existence of the cellulosic and resinic compounds in coal, the primary decomposition of coal substance begins at 200° C; with elimination of water and oxides of carbon, up to and beyond 450° C; hydrogen continues to evolve until at 700°—800° C it suddenly increases in volume. Burgeas and Wheeler regarded this temperature as the critical temperature at which hydrogen-yielding constituents are decomposed while other investigators considered the sudden increase in volume of hydrogen as indicating the secondary composition of tar vapors or gaseous hydrocarbons. Aromatic hydrocarbons of the benzene series are not found in low temperature carbonisation. Perhaps the benzene series found at higher

temperature are due to the dehydrogenation of the hydroaromatic products found at low temperature or due to olefine condensation. So far there has been no systematic investigation of the modes of decomposition of ammonia-yielding nitrogenous constituents.

IV. PHOTOGRAPHIC.

W. T. Russell discovered long ago that coal has the property of acting on a sensitized plate in the dark. This kind of investigation has been recently resuscitated by G. S. Haslam and R. V. Wheeler who observed that the contact photograph recorded with remarkable fidelity the slightest variation in the texture of coal and showed up clearly the bands of vitrain, clarain, fusain and durain. I am of the opinion that this method would eventually prove to be the quickest method for the investigation of the four ingredients of coal.

V. X-RAY EXAMINATION.

This method of research has been used by Chôzo Iwasaki in his investigation of Japanese coals. I have not been able to find any allusion to it in European or American literature. Perhaps it would be a good method for the investigation of the mineral content of coal.

VI. CONCLUSION.

In the space given to this paper, I have tried to draw your attention to the lines of attack for the investigation of the constitution of coal, which have been adopted by various researches. This summary is to give you a general view of the results so far obtained, to show you the relationship such researches would bear to the work of correlating coal seams, and to inform you how incomplete our present knowledge of the constitution of coal is. However, I hope that, with the vigorous investigations now carried on by many arduous workers in different lands, the day would not be far off when we shall be able to write down chemical formulæ for the coal mass as do now we can do with a mineral. May we not now in this old land of Sinian, which possesses such an immense wealth of coal, try to add our mite of work for the unravelling of the mysteries of the constitution of coal?

In conclusion, may I be permitted to suggest to you that the Geological Society should take the lead in the establishment of a central organiza-

tion for the scientific study of Chinese coal?

SOME RECENT IMPORTANT REFERENCES.

1) *Books and pamphlets:*

- Bone, William A. *Coal and its Scientific uses*, 1918.
- Fisher, Franz and Schrader, Hans. *Entstehung und chemische Struktur der Kohle. Aus dem Kaiser-Wilhelm-Institut für Kohlenforschung in Mülheim Rube*, 1922.
- Robertson, Tohu B. *Chemistry of Coal*, 1919.
- Sinvatt, F. S., Stern, H. and Bayley, F. *Coal Dust and Fusain. The Lancashire and Cheshire Coal Research Association, Bull. 5*, 1920.
- Stopes, M. C. and Wheeler, R. V. *The Constitution of Coal. Dept. of Scient. and Ind. Research*, 1918.
- Thiessen, Reinhardt. *Structure in Palaeozoic bituminous coals. Bull. 117, Bureau of Mines, U. S.*, 1920.
- White, David and Thiessen, R. *The Origin of Coal. Bull. 38, Bureau of Mines, U. S.*, 1913.
- White, David. *Resins in Palaeozoic plants and in coals of high rank. Prof. Paper 85, U. S. G. S.*, 1914.

2) *Papers:*

- W. A. Bone, A. R. Pearson, E. Sinkinson, and W. E. Stockings. *Researches on the chemistry of Coal. Pt. II. The resinic constituents and coking properties of coals. Proc. Roy. Soc. A*, 100, pp. 582—598, 1922.
- Findley, A. E. and Wigginton, R. *The separation of the constituents of banded bituminous coal. Fuel in Sci. and Pract. June 23*, 1922, pp. 106—107.
- Grounds, A. *A contribution to the study of the constitution of anthracite. Jour. Soc. Chem. Ind.* 1922, pp. 88-92 T.
- Haslam, G. S. and Wheeler, R. V. *Contact Photographs of coal. Fuel in Sci. and Pract. March 1922*, pp. 43—47.
- Hichling, G. *Micro-petrology of coal. Trans. Inst. Min. Eng. May*, 1917.
- Illingworth, S. Roy. *Coal and its carbonisation. Fuel in Sci. and Pract. Feb. 1922*, pp. 17—19.

- Illingworth S. Roy. Researches on Coal. Jour. Soc. Chem. Ind. May 15, 1920, pp. 111 T—118 T; May 31, 1920, pp. 133 T—138 T.
- Iwasaki, Chozo. A fundamental study of Japanese coal. Technology Report of Tohoku Imp. Uni. No. 2. Reprint: Coll. Guard. Sept. 17, 1920, pp. 797—799; Oct. 1, pp. 941—942; Oct. 8, pp. 1027—1028; Oct. 15, pp. 1088—1089.
- Jones, D. T. and Wheeler, R. V. On the formation and chemical constitution of coal. Fuel in Sci. and Pract. June 23, 1922, pp. 91-93.
- Jones, D. T. and Wheeler R. V. The composition of coal. Trans. Chem. Soc. Lond., Vol. 105, 1914, pp. 140—151; Vol. 105, 1914, pp. 2562—2565; Vol. 107, 1915, pp. 1318—1324; Vol. 109, Vol. 1916; pp. 707—714.
- Lessing, R. The Constitution of Coal. The Gas World, coking section. Aug. 5, 1922, pp. 11—13.
- Lessing, R. The Carbonisation of Coal. Fuel in Sci. and Pract. Aug. 2, 1922, pp. 137—149.
- Lomax, T. The formation of coal-seams in the light of recent microscopic investigations. Trans. Inst. Min. Eng. Vol. 50, 1915, pp. 127—158.
- Lomax, T. The preparation of transparent section of coal. Fuel In Sci. and Pract. May, 1922, pp. 79—84.
- Porter, H. C. and Taylor, G. B. On low temperature distillation of coal. Proc. Amer. Gas. Inst. 1914, pp. 234—238.
- Reim, E. P. The combustion of south African Coals in Boiler Furnaces. Jour. Chem. Met. Min. Soc. South Africa. Vol. 22, 1922, pp. 117—134.
- Sinvatt, F. S. A method of representing the structure of coal-seams, and the proportion and properties of the four constituents (vitrain, clarain, durain, fusain) contained in certain seams. Trans. Inst. Min. Eng. 1922, pp. 307—317.
- Sinvatt, F. S. The constitution of a coal seam. Iron and Coal trades review Dec. 23, 1921, p. 912.
- Stopes, M. C. The Constitution of coal. Fuel in Sci. and Pract. June 23, 1922, pp. 93-101.
- Stopes, M. C. Remarks on Vitrain Fuel in Sci. and Pract. Feb. 1922, pp. 22—25.

- Thiessen, R. Compilation and composition of Bituminous coal. *Journal of Geol.* April—May, 1920, pp. 185—209.
- Thiessen, R. Recent developments in the microscopic study of coal. *The Coal Mining Quart. of America.* 1920, pp. 88—120. Also coal age, Dec. 9, 1920, pp. 1183—1189; Dec. 16, pp. 1223—1227; Dec. 23, pp. 1275—1279; Jan. 6, 1921, pp. 12—15.
- Wheeler R. V. and Wigginton. Resins in bituminous coal. *Fuel in Sci. and Pract.* Jan. 1922, pp. 10—14.