THE RECTANGULAR GRAPHS AS APPLIED TO THE PROXIMATE ANALYSES OF CHINESE COALS.

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WITH ONE PLATE AND TWO FIGRUES.

I. INTRODUCTION.

On interpretation of the chemical analyses of any substance, it is often better to set forth the data on a suitable method of graphic representation than to study them merely by long columns of figures in order to express more clearly their various points of resemblance or difference, their relationships or other characters of no less importance. Since the time of Prof. Parr[1] and Newberry[2], various kinds of graphs or graphic representations have been devised from time to time for expressing the coal composition, among which we may especially mention the ultimate-analysis triangular diagram of Grout[3] and Ralston[4], the Campbells'[5] coal-rank diagram of proximate analysis and more recently the proximate-analysis triangular diagram of Dr. Wong[6] and Fisher[7]; each apparently has its own peculiar merits.

But apart from Dr. Wong[8] who has discussed in a broad way the relation between the moisture content and the geological age, not enough attention has been so far paid to the bearing of the geological age upon the coal composition. As coals vary very widely in ages, from late Palæozoic to Tertiary, and as revealed already by Palæobotany, each geological time has its

1. Universities of Illinois studies, Vol. 1, No. 7, Illinois Geological Survey, Bull. 3, p. 33.
2. Economic Geology by Heinrich Ries (1911), p. 12.
3. "The Composition of Coals", Economic Geol. Vol. II (1907), p. 227.
4. "Graphic studies of Ultimate Analyses of coals," U. S. Bureau of Mines Tech. Paper 93 (1915).
5. "The Coal Fields of The United States" U. S. Geol. Survey Prof. Paper 100-A (1922), p. 8.
6. "Coal Composition in Triangular Diagram"; Bull. Geol. Soc. China, Vol. 6, No. 1, (1927), p. 66.
7. "Notes regarding the Coalification Process", Journal of Geology, Vol. XXXV, No. 7, University of Chicago (1927), p. 640.
8. "Classification of Chinese Coals and a New Nomenclature with Notations". Oriental Engineer, (1926), p. 19. and Bull. Geol. Surv. China No. 8 (1927), pp. 40-52.

own peculier flora from which is solely derived the coal substance, it is to be expected that different flora might have different composition which, in turn, have cast different shadows upon the coals constituents. However, no such relation has been clearly made out, up to the present, by any one of the above mentioned diagrams.

The graphic representation here proposed is primarily introduced to study the chemical changes of Chinese coals in the process of coalification. It gives two separate curves for both volatile matter and moisture, which correspond respectively to coals of two different era, namely the Palæozoic and the Mesozoic. It, therefore, brings out what has been overlooked by the above diagrams and is perhaps not unworthy of record.

Though the proximate analysis alone does not satisfactorily express the variations in coal, yet the data of the ultimate analyses for Chinese coals are so scanty that it is not yet sufficient for consideration. This inevitably forces the author to consider, instead of both, only the former, namely the proximate analysis.

In constructing the diagram, only three constituents, namely the fixed carbon, the volatile matter and lastly the moisture, all on ash-free basis as calculated from the proximate analysis, are used. It is somewhat analogous in principle to one of the graphs of Grout,[1] as both use the fixed carbon as the abscissa, but differs from the latter in that it takes, instead of the total carbon, both volatile matter and moisture on the ordinate. Plate I is an example so constructed from the 40 analyses of Chinese coals. The reasons why we should use the moisture as well as the volatile matter in exclusion of the ash content are two-fold: firstly because the mineral matter originally contained in the vegetatin is usually so small that it only constitutes a trifling portion of the ash while its remaining essential part is either sandy or clayey material of a fortuitous origin; secondly, because the moisture, though of minor importance in high-rank coals, is present in relatively large amount in the low-ranks, and therefore must be taken into consideration as has been pointed out by Ashley[2] and Thom[3].

The data used is essentially the same as that compiled by Dr. Wong[4] with some new analyses recently made by Messers. Liu and King of the Geological Survey. They are tabulated in the following table with their respective recalculated ash-free contents and Calorific values.

1. Grout, F. F. op. cit. p. 237.
2. "A Use Classification of Coal", Trans. A. I. M. E., Vol. LXIII (1920), p. 783.
3. "Moisture as a Component of the Volatile Matter of Coal", Trans. A. I. M. E., Vol. LXXI (1925), pp. 282-288.
4. Wong, W. H., op. cit. pp. 2—3.

TABLE OF PROXIMATE ANALYSES OF PRINCIPAL KINDS OF CHINESE COAL WITH THEIR RESPECTIVE THREE RECALCULATED ASH-FREE COMPONENTS AND CALORIFIC VALUES.

No.	Province	Coal mine	Proximate Analyses.				Ash-free Basis				Geological Age.
			M.	V.M.	F.C.	A.	M.	V.	F.C.	C.V.	
1.	Hopei (Chihli)	Kaiping	0.60	25.98	59.78	13.34	0.74	30.00	69.00	7,443	Permo-Carboniferous
2.	,,	Lincheng	1.89	30.88	56.64	11.50	2.10	34.50	63.10	7,529	,,
3.	,,	Chinghsing	0.56	20.20	69.20	9.20	0.62	22.50	77.00	7,813	,,
4.	,,	Liukiang	0.70	6.57	77.57	15.20	0.80	7.76	91.50	7,259	,,
5.	Shansi	Pingting.	0.78	6.55	86.35	5.70	0.83	7.00	92.10	7,991	,,
6.	Honan.	Chiaotso	0.65	6.70	84.50	8.00	0.70	7.30	92.00	7,854	,,
7.	,,	Liuhokou	0.55	19.16	72.05	8.40	0.60	20.80	78.90	7,976	,,
8.	Shantung	Changchiu	0.75	13.30	54.36	31.60	1.09	19.80	79.30	5,913	,,
9.	,,	,,	0.68	15.07	66 96	17.29	0.83	18.20	80.80	7,175	,,
10.	,,	,,	0.45	17.37	55.51	26.67	0.62	24.10	75.50	6,357	,,
11.	,,	,,	0.62	12.62	49.51	37.25	0.99	20.10	78.90	5,424	,,
12.	,,	,,	0.53	13.32	71.27	14.89	0.62	15.60	83.70	7,386	,,
13.	,,	,,	0.57	13.19	74.98	11.27	0.64	14.80	84.20	7,697	,,
14.	,,	Chunghsing	0.50	27.00	63.50	9.40	0.61	29.60	69.70	7,858	,,
15.	,,	Poshan	0.85	18.90	69.80	9.85	0.95	21.20	78 00	7,755	,,
16.	,,	Tzechuan	0.57	14.90	74.70	10.00	0.63	16.50	82.80	7,833	,,
17.	,,	Paohua	1.44	27.89	62.27	8.42	1.57	30.40	68.10	7,823	,,
18.	,,	,,	1.67	36.97	56.96	4.40	1.75	38.60	59.20	7,672	,,
19.	,,	,,	2.08	33.80	54.91	9.21	2.30	37.20	60.20	7,368	,,
20.	,,	,,	1.98	35.01	56.42	6.59	2.11	37.30	60.10	7,586	,,
21.	,,	,,	2.49	33.31	59.35	4.85	2.61	35.00	62.20	7,898	,,
22.	Fengtien	Penchihu	0.68	23.95	64.07	11.20	0.77	27 00	72.20	7,668	,,
23.	,,	Wuhutzui	0.83	8.60	85.31	5.26	0.88	9.06	90.10	8,160	,,
24.	,,	,,	0.65	8.97	81.46	8.92	0.71	9.82	89.40	7,852	,,
25.	,,	Yentai	1.15	12.00	71.20	14.70	1.30	14.20	84.70	7,262	,,
26.	Anhuei	Hsuancheng	0.75	24 47	52.65	22.00	0.97	31.40	67.50	6,677	,,
27.	,,	Shunkengshan	0.85	34.90	54.75	9.50	0.94	38.50	60.20	7,370	,,
28.	Kiangsu	Chiawang	1.35	29.54	51.50	17.60	1.64	35.90	62.40	6,873	,,
29.	Chekiang	Changhsing	0.94	37.70	49.80	10.90	1.60	42.40	56.00	6,859	,,
30.	Hopei (Chihli)	Mentoukou	2 30	6.50	75.20	15.00	2.73	7.71	89.50	7,050	Jurassic
31.	Jehol	Peipiao	3.25	30.50	54.25	11.00	3.70	34.60	62.00	7,219	,,
32.	,,	Hsinglungkou	0.76	27.13	60.42	11.69	0.83	60.70	68.00	7,594	,,
33.	Shansi	Tatung	4.50	30.99	59.45	5.50	4.70	32.60	62.80	7,725	,,
34.	Kiangsi	Pinghsiang	1.35	23.75	62.75	11.80	1.54	27.10	71.40	7,531	,,
35.	Honan	Mienchih	9.60	32.50	45.40	13.50	10.98	37.20	51.90	6,178	,,
36.	Shantung	Fangtzu	2.80	30.70	51.80	14.70	3.28	36.00	60.50	6,928	,,
37.	Kirin.	Yütung	15.61	36.37	46.50	11.46	15.82	36.80	47.15	6,460	,,
38.	Heilungkiang	Zalainor	20.93	36.35	39.03	3.69	21.70	37.80	40.60	5,741	Tertiary
39.	,,	,,	21.50	35.50	38.00	5.00	22.60	37.40	40.00	5,591	,,
40.	Fengtien	Fushun	6.73	39 34	48.15	5.25	7.12	41.60	51.10	7,148	,,

M. representing the moisture; V. M. the volatile matter; F. C. the fixed carbon; A. the ash content; C. V. the Calorific value.

II. Explanation of the Diagram

A glance at Pl. I. will reveal the following facts which are clearly indicated:

1. One of the most remarkable features is the continuity as well as the smoothness of the curves of the volatile matter, and this is of special note when the scale of the graph, say 4 mm for one percent, is considered. It then signifies clearly the fact that there is no break in the series from low-rank bituminous to anthracite in Palæozoic coals and in those of both Mesozoic and Tertiary. Therefore, whatever may be the supposition of Donath[1] who has denied the derivation of coal from lignite, it seems fairly evident, so far as this diagram reveals, that coal is derived by a series of changes, from vegetable matter, through peat, lignite, soft coals to anthracite, forming a series of substances which grade one into another in an unbroken line. Such a gradual chemical change has long been noted and more recently, Dr. Wong has plotted some analyses of Chinese coal on a triangular diagram which will bring out the same relation, if slightly modified. Nevertheless, for clearness as well as for

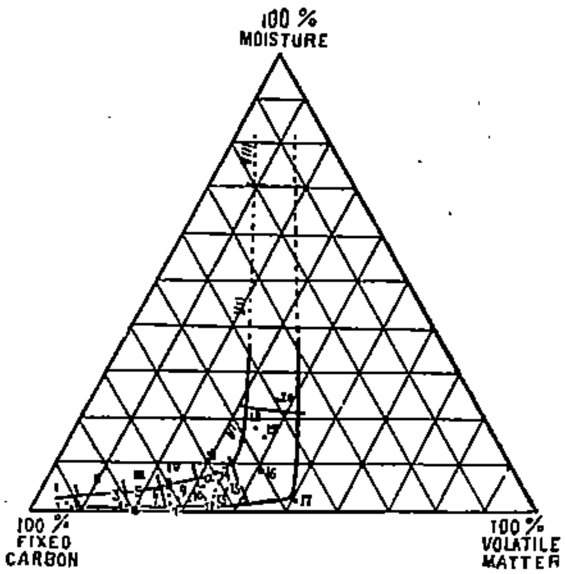

Fig. 1. Triangular diagram to show the continuous series of 20 Chinese coals in terms of proximate-analyses constituents: I Ah; II, AB; III, Bh; IV, Bm; V, Bl; VI, BC; VII, C; VIII, Peat. (Modified after Dr. Wong).

ready grasping at a mere glance, the former, namely, the rectangular graph seems to be much better than the latter. The absence of lignite in the

1 Zeitschr. anorg. Chemie (1906), p. 657.

Palæozoic series is perhaps due to the fact that time also plays an important rôle in coal metamorphism and, consequently, all coal deposits of Palæozoic time would be more profoundly metamorphosed on account of the long duration.

2. It shows remarkable differences in chemical transformations during the different stages of coalification. For instance, the curve of the moisture gradually but rather steeply descends from lignite to low-rank bitumite while that of the volatile matter is more or less horizontal or only slightly slopes downward. This indicates that dehydration is of overwhelming importance during this first stage, from lignite down, with only a slight devolatilization. From the low-rank to the medium-rank bitumite, both dehydration and devolatilization go hand in hand, though the latter becomes more and more marked. From the medium rank bitumite up to the anthracite, instead of dehydration, devolatilization is the previaling process. Throughout all these stages the amount of fixed carbon increases steadily, and is, therefore, the most prominent feature among all the chemical changes. This accounts for why, in this diagram, we use it as the abscissa rather than the volatile matter or the moisture content.

However, there are few exceptions: for example, the sudden rise of the moisture for both Tatung (33) and Peipiao (31) coals, the irregular undulations between the interval from low-rank bitumite to semi-anthracite, and the high volatile-matter content of Fushun coal. Some reasonable explanation for these special features may be found by future research. But the lack of standardization of sampling, and analyzing of these samples considered, would perhaps explain many of these apparent irregularities.

3. There are two pairs of curves as differentiated on the diagram respectively for both volatile matter and moisture. In each pair one curve represents the Palæozoic coals and the other embodies those of the Mesozoic. Granting this fact, it, therefore, brings out the general difference in composition between the Palæozoic and the Mesozoic coals. The former is comparatively higher in volatile matter and lower in moisture while the reverse is true for the latter. This may be due to the original difference in composition between the Palæozoic and the Mesozoic flora resulting from the great diastrophism at the end of Palæozoic time.

4. Two curves of the Calorific values which are calculated from Goutals[1] method are also given. The higher one corresponds to those with less ashy content, while the lower to those containing preponderant ash. They show that the highest Calorific value is not for the last stage in the

1. De la houille (1921), pp. 68-70.

metamorphism, namely the Pingting anthracite in this case, but with that of Wuhutzui (No. 23) and show further that of the Liuhokou high-rank bitumite nearly attains the same figure as that of the Pingting Coal. As Campbell[1] express, "The greater heating power of the low-rank coals as compared with anthracite is due to the fact that these coals contain a considerable quantity of available hydrogen which when burned produces a much greater heat than the same weight of carbon." Further, the sudden rise and abrupt fall of these curves is of special note indicating that there must be some other factors which should be taken into account when the Calorific value is discussed. The most important ones are, in all probability, the ash and moisture content, one of which, namely the ash, has been purposefully omitted in constructing the graph. This shows the necessity of the indication of *Grades* of coal, in addition to *Ranks*, in the new nomenclature of Dr. Wong.

In order to express more clearly the chemical changes above described the following figure is constructed (Fig. 2).

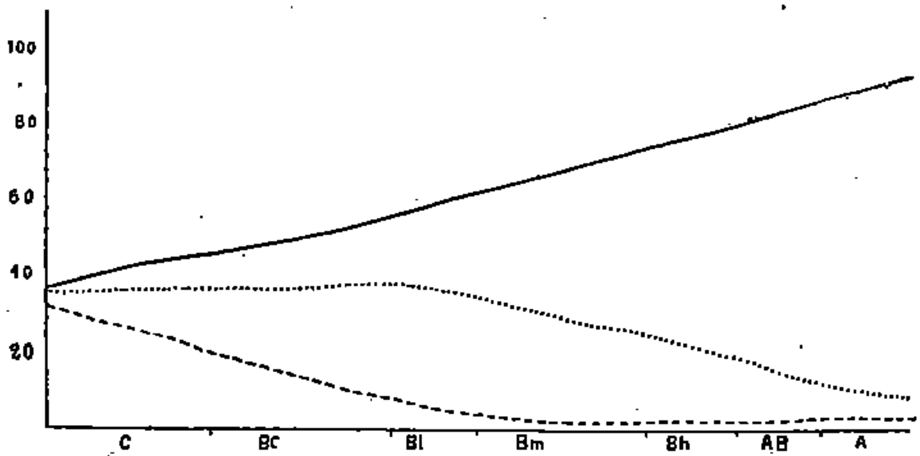

Fig. 2. --Diagram to show the relative change in amount of the three constituents, the fixed carbon (solid line), the volatile matter, (dotted line) and the moisture (dashed line) during the coalification process. From lignite (c) passing through lignitic bitumite (BC) to low rank bitumite (Bl) an increase of fixed carbon with a decrease of moisture; from the low-rank bituminous stage to the medium rank both decrease of volatile matter and of moisture accompanied with an increase of fixed carbon, and thence onward, a decrease of the volatile matter and an increase of the fixed carbon constitute the essential features. Only during the last two stages, namely, the anthracitic bitumite (AB) and the anthracite, (A), the moisture tends to increase in a slight amount.

It is rather self-explanatory and so needs no further explanation.

1. Campbell M. R. op. cit. p. 9

III. THE CLASSIFICATION OF THE COALS.

Granting the facts as revealed by this diagram, and noting the continuity as well as the smoothness of the curves, it seems to the author that there is an equally perfect soundness between the classification of coals upon the percentage of fixed carbon on an ash-free basis and the Ashley's method which has been adopted by Dr. Wong for classifying the Chinese coals. The following table is tabulated from both methods for comparison. Their remarkable correspondence in order with only a few exceptions is of special note.

TABLE OF CLASSIFICATION OF THE CHINESE COALS BASED ON THE FIXED CARBON OF THE ASH-FREE BASIS.

Order According to Ash-free fixed carbon	Order as given by Dr. Wong According to Ashley's method	Coal.	Ash-free fixed carbon	Symbol.
1.	1 (11.80)	Pingting	92.10	Ah_3
2.	2 (11.50)	Chiaotso	92.00	Ah_2
3.	3 (10.50)	Liukiang.	91.50	Ah_4
4.	4 (9.10)	Wuhutsui	90.10	Am_1
5.	5 (8.50)	Mentoukou	89.50	Am_4
6.	6 (8.50)	Wuhutsui	89.40	Am_2
7.	8 (5.40)	Yentai	84.70	AB_4
8.	7 (5.50)	Changchiu	84.20	AB_2
9.	9 (5.20)	,,	83.70	AB_4
10.	10 (4.90)	Tzuchuan	82.80	AB_3
11.	11 (4.30)	Changchiu	80.80	AB_4
12.	12 (3.86)	,,	79.30	Bh_2
13.	13 (3.74)	,,	78.90	Bh_2
14.	14 (3.56)	Liuhokou	78.90	Bh_3
15.	15 (3.54)	Poshan	78.00	Bh_2
16.	16 (3.40)	Chinghsing	77.00	Bh_1
17.	17 (3.12)	Changchiu	75.50	Bh_2
18.	18 (2.60)	Penchihu	72.20	Bm_1
19.	19 (2.50)	Pinghsiang	71.40	Bm_3
20.	20 (2.30)	Chunghsiang	69.70	Bm_2
21.	21 (2.20)	Kaiping	69.00	Bm_4

22.	22 (2.11)	Paohua	68.10	Bm_3
23.	23 (2.16)	Hsinglungkou	68.00	Bm_2
24.	24 (2.10)	Hsuancheng	67.50	Bm_5
25.	25 (1.70)	Lincheng	63.10	Bm_1
26.	26 (1.67)	Tatung	62.80	Bl_9
27.	27 (1.66)	Chiawang	62.40	Bl_4
28.	28 (1.66)	Paohua	62 20	Bl_8
29.	29 (1.60)	Peipiao	62.00	Bl_3
30.	32 (1.50)	Fantzu	60.50	Bl_4
31.	30 (1.58)	Paohua	60.20	Bl_2
32.	33 (1.50)	Shunkengshan	60.20	Bl_7
33.	31 (1.55)	Paohua	60.10	Bl_9
34.	34 (1.47)	Paohua	59.20	Bl_3
35.	35 (1.30)	Changhsing	56.00	Bl_1
36.	36 (1.10)	Mienchih	51.90	BC_4
37.	37 (1.00)	Fushun	51.10	BC_3
38.	38 (0.90)	Yütung	47.15	BC_1
39.	39 (0.70)	Zalainor	40.60	C_1
40.	40 (0.67)	,,	40.00	C_2

And when it happens that two samples or more than two possess the same percent of fixed carbon but with different amounts of volatile matter, for example as No. 13, 14 and No 15, 32, we use the ratio of fixed carbon to volatile matter as a criterion for determining their positions in the series. The larger this ratio the higher will be the order of the sample, considered, in the series. This places No. 14 below No. 13, and No. 32 below No. 31.

However, it must be remarked here that such a kind of graph is only introductory for studying some problems concerning the coal, and as only a few analyses are used in this paper, it is still hazardous to generalize too much.

IV. PHYSICAL ACTION IN THE PROCESS OF COALIFICATION.

Besides the chemical changes physical agents may have played an important rôle in the process of coalification. Such physical action has been in fact proved by artificial experiments. There have been two lines of attack upon this problem. In the first instance, some scientists sought to produce artificial coal from wood or some other vegetable matter solely by means of pressure, and among them we should mention W. Spring[1], R Zeiller[2] and Gumbel[3].

1. Bull. Acad. roy. Sci. Belgique, Vol. 49, (1880), p. 367.
2. Bull. Soc. Géol. France, 3d Ser., Vol. 12 (1884) p. 680.
3. Sitzungsb. Math-phys. Classe. K. bayer. Akad. Wiss. München, Vol, 13 (1883), p. 141.

In the second instance, heat was used as the sole potent transforming agent and this has been repeatedly carried out by Latour[1], Violette[2], Fremy[3] and S. Stein[4] since the time of Sir James Hall. What we can infer from these experiments is that heat is more important in the process of coalification than pressure but the latter also plays a not negligible rôle for, on one hand, Gumbel once subjected lignite to pressure as high as 20,000 atmospheres but with no serious change in it and, on the other, Violette has prepared from wood a fatty coal in a sealed tube heated to 400°C under pressure, while charcoal has no likeness to any kind of coal on account of its free from pressure in the kiln. The function of heat is to decompose the organic matter and that of pressure is to retard the change and to prevent the escape of the volatile product. But even through the combined effect of both heat and pressure, the similarity between the artificial and the natural coal is not at all complete. This is why most authorities agree in the belief that both the microorganism as well as the factor of time must be taken into account when the mechanism of coalification is concerned. The present paper on Chinese coals seems fully to confirm the importance of geologic age in relation to the coal classification.

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1. Compt. Rend., Vol. 32, (1851), p. 295.
2. Annales Chim. phys., 3d Ser., Vol. 32, (1851), p. 304.
3. Compt. Rend. Vol. 88, (1879), p. 1048.
4. Chem. Centralbl. (1901), pt. 2, p. 950.