SOME PROBLEMS RELATING TO SOUTH AFRICAN COKING COALS AND THEIR BEHAVIOUR DURING CARBONISATION.

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INTRODUCTION.

One of the main problems of the carbonisation industry in South Africa is the limited choice of coking coals suitable for carbonisation purposes.

The existing coking coal reserves can be described as consisting of medium ash, high volatile coal. The proximate and ultimate analyses of a number of the more important coking coals are given in Table 1.

TABLE 1.
Proximate and Ultimate Analyses of Some South African Coking Coals.

Colliery	Proximate Analyses(%)(air-dry)					Ultimate Anal.(%)(D.A.F.)			
	H_2O	Ash	Vol. Mat	Tot.S	Sw. No.	C	H	O	Vol. Mat.
A (Transvaal)	2.7	10.4	32.9	0.7	3	83.0	5.3	8.9	38.1
B (")	2.6	10.3	34.0	0.7	3½	83.5	5.4	8.9	39.0
C (")	2.2	13.1	30.9	0.5	3	84.0	5.1	8.5	37.0
D (Natal)	1.3	12.0	31.0	1.4	6	85.5	5.3	6.0	37.5
E (")	0.9	13.7	22.9	1.5	7	88.1	5.0	3.7	27.1
F (")	1.1	12.5	22.9	0.9	4	88.3	4.8	4.3	26.9
G (")	1.1	14.9	20.8	0.8	4½	88.5	4.7	4.2	24.2

Practically all coals intended for carbonisation are mechanically cleaned to yield a product with an ash content of about 12 per cent. The Transvaal coals used for this purpose are only weakly coking. They yield a very inferior product when carbonised on their own, but acceptable coke can be produced by blending with a small percentage of certain Natal coking coals. These are of a slightly higher rank as the result of the maturing effect of intrusive dolerite sheets some distance either below or above the coal horizon.

Economic considerations and the desire to conserve coking coal reserves have determined the current policy whereby most of the metallurgical coke in South Africa is produced from blends having weakly coking coal as main component. The total tonnage of such blends carbonised during 1959 was 2.2 million tons* representing nearly 70% of all coal carbonised for the production of metallurgical coke in this country.

A typical blend is:

Weakly coking coal: (usually a mixture of various weakly coking coals)	75%
Coking coal (normally a mixture of coking coals).	25%

A. THE PETROGRAPHIC COMPOSITION OF PREPARED COKING COALS.

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* Short tons of 2,000 lb.

The South African coking coals are characterised by the presence of very large amounts of inert material. For the microscopical examination of these coals the incident light technique has therefore been found preferable to the transmitted light technique currently used in the U.S.A.[1)]

The petrographical analyses of some South African coking coals are given in Table 2.

TABLE 2.

Petrographic Analyses of Some South African Coals.

Colliery	Microlithotype Analysis (%)						Maceral Analysis(%)			
	Vt	Cl	V.I.	I.M.	Fu	C.S.	Vn	Ex	In	V.M.
A(Transvaal)	27.5	6.0	23.0	38.5	0.5	4.5	55.8	14.4	25.0	4.8
C(")	19.5	7.5	31.4	39.6	0.3	1.7	47.9	12.0	35.2	4.9
D (Natal)	26.9	4.0	33.3	30.2	1.3	4.3	51.9	9.3	33.1	5.7
E (")	20.9	2.7	30.7	38.7	2.2	5.8	51.0	10.4	31.2	7.4
F (")	16.2	2.7	30.9	42.1	2.7	5.4	42.3	14.0	37.7	6.0
H (")	7.8	-	53.3	36.0	0.2	2.7	45.0	12.4	39.8	2.0

Exinite is rarely found to be associated with the vitrinite. Only relatively small quantities of clarite are therefore present in the coal. Normally the clarite content varies between 2 and 7 per cent in the coking coals and any value higher than 7 per cent can be regarded as exceptional. The small amount of exinite present in the coal (usually less than 20 per cent) is found associated with mixtures of vitrinite and inertinite. This gives rise to large amounts of claro-durite and duro-clarite, generally described as intermediate or transition material.

Vitrinite in its free form i.e. in bands, is fairly common but the bands are rather thin and the vitrite content seldom exceeds more than 25 to 27 per cent in a microlithotype analysis. The normal amount is of the order of 20 per cent. At least half of the total amount of vitrinite occurs admixed with other macerals.

Another very common entity is the microlithotype vitrinertite which consists of an intimate mixture of inertinite (mainly semi-fusinite) and vitrinite. This microlithotype is usually very consistent in composition and contains few or practically no other macerals. It occurs in very large quantities in some South African coals, notably the non-coking coals, as much as 60 per cent having been recorded in certain instances. For practical purposes this microlithotype is recorded separately whereas durite, which occurs only in small quantities (seldom more than 8 per cent), is grouped and determined together with the intermediate material.

Fusite occurs sparsely and is generally highly mineralised. The friable variety is rare.

Since virtually all the coals intended for high temperature carbonisation are mechanically cleaned the amount of carbonaceous shale is relatively low in them. Minute nodules of clay which cannot be separated from the coal by normal mechanical cleaning processes are associated with practically every microlithotype, even in comparatively low specific gravity fractions. These microscopical nodules of clay give rise to the typical high ash content of the South African coals.

B. .../

B. THE COMPOSITION OF SPECIFIC GRAVITY FRACTIONS.

A study of the specific gravity distribution of the microlithotypes in -22 mesh coal grains of a weakly coking coal and of two coking coals revealed a remarkable similarity in the behaviour of the microlithotypes. As can be expected, the largest concentration of vitrite was found in the float material at 1.30 s.g. Values ranging from 50 to 60 per cent were obtained for the coking coals and just over 20 per cent for the weakly coking coal. The vitrite concentration in both types decreased rapidly with increasing specific gravity so that the 1.35-1.40 s.g. fraction contained only 6 - 7 per cent vitrite. It then falls very gradually reaching a value of about 2 per cent in the 1.70 s.g. sink material.

A similar distribution pattern was observed for clarite in the weakly coking coal. Here the maximum concentration of clarite was found in the 1.30 s.g. float material viz. 24.3 per cent. It fell to 3 per cent in the 1.30-1.35 s.g. fraction and no clarite was found in the higher specific gravity fractions. The 1.30 s.g. float material from the two coking coals contained no more than 6 - 7 per cent clarite. The concentration decreased fairly steadily with increasing specific gravity reaching a value of about 1 per cent in the 1.70 s.g. sink material.

The behaviour of the intermediate material was similar to that of the vitrite. The maximum concentration in the 1.30 s.g. float was appreciably higher for the weakly coking coal than for the two coking coals.

The concentration of the vitrinertite in the 1.30 s.g. float material amounted to about 20 per cent for the coking coals and less than 10 per cent for the weakly coking coal. It then increased rapidly for all three coals, attained a maximum of between 70 and 80 per cent in the specific gravity range 1.40-1.55 and decreased rapidly to 10 per cent in the 1.70 s.g. sink fraction.

The concentration of the fusite remained practically constant for all these coals through the specific gravity range and seldom exceeded 5 per cent.

No carbonaceous shale was present in any of the three 1.30 specific gravity floats. It appeared in concentrations of 1 to 2 per cent in the 1.30 - 1.35 specific gravity fractions and thereafter the concentration steadily increased with increasing specific gravity until it finally reached a maximum of between 80 and 90 per cent in the 1.70 specific gravity sink material.

From the study of the specific gravity distribution of the microlithotypes in the coals it appears: (1) That the microlithotypes are very intimately mixed. Thus with very few exceptions, practically every microlithotype was represented in the whole range of specific gravity fractions. (2) That the specific gravities of the microlithotypes are appreciably higher than those of European coals of the same rank. The values, statistically determined, varied within the following limits:

Vitrite	1.31-1.33
Clarite	1.30-1.32
Vitrinertite	1.47-1.49
Intermediate material	1.38-1.40
Fusite	1.44-1.54
Carbonaceous shale	1.63-1.65

The possibility of contamination of the microlithotypes by quantities of clay, so minute that they could not always be observed under normal magnification of 200 to 250, may have influenced these specific gravities although very little clay was detected under normal

magnification .../

magnification in the samples of specific gravity fraction up to 1.50. It is significant, however, that only slight variations in specific gravity occurred for each microlithotype for all three of the coals. The exception being the highly mineralised fusite in the weakly coking coal which gave an appreciably higher value than that obtained for the fusite in the two coking coals. The coals originated from areas very widely apart and it is doubtful whether such constant results could have been obtained if the inclusion of minute quantities of clay had had any profound influence on the specific gravity.

C. THE EVALUATION OF COKING COALS.

A prediction of the potential value of South African coking coals, more specially that of weakly coking coals, is frequently rendered rather difficult by their peculiar behaviour.

Thus the widely used B.S.swelling test does not provide an entirely reliable criterion. From past experience with South African coking coals it appeared that coal having a swelling number of less than 2 was of very doubtful value as a coking coal although some of the coals having such a low swelling number can be used in the blends mentioned above. It was usually accepted that these weakly coking coals should have a swelling number of 2 to 4. A higher swelling number provides no guarantee that the coal will yield satisfactory coke.

The fallibility of this test became apparent when specific gravity fractions from 1.30 to 1.60 at 0.05 intervals of a reasonably good Natal coking coal were tested in a dilatometer. The dilatation of the coal became progressively poorer with increasing separation specific gravity until the float coal finally gave only a contraction; yet the swelling number varied from 6½ in the case of the lowest specific gravity fraction, to 4½ in the case of the highest s.g.fraction.

An evaluation on the basis of petrographic composition, provided the coal falls within the coking group for South African coals, also offers considerable difficulties. It has been found that the ratio of active to inert constituents obtained from the maceral analysis may give some indication of the coal's coking propensity. If the ratio of the sum of the vitrinite and exinite to that of the inertinite and the visible minerals exceeds 1.4:1, the coal may exhibit reasonable coking properties. Coals having ratios of actives to inerts of more than 2:1 can be expected to give a reasonably good coke when carbonised.

Extensive testing of coking coals using the dilatometers of Audibert-Arnu[2] and Hoffmann[3], as well as the Gieseler[4] plastometer indicate that a reasonable contraction, dilatation and degree of fluidity could be expected from most of the Natal coking coals. Normally coal D (Table 1) gives 22 to 25 per cent contraction, 30 to 40 per cent dilatation and a maximum fluidity of about 3000 div./min. when tested by itself. The behaviour of the Transvaal weakly coking coals is at times difficult to predict from such tests, especially when they are to be blended with Natal coking coals. Under test conditions the Transvaal weakly coking coals exhibit remarkably similar properties. They all give a contraction but no dilatation. In the plastometer their degree of fluidity is very low. A blend of 30 per cent of coal D from Natal with 70 per cent of coal A from Transvaal gave a contraction in the dilatometer of 32 per cent and a dilatation of -28 per cent. The degree of fluidity was low with a maximum of 14 div./min. If 5 per cent anthracite is added to coal E from Natal the maximum fluidity drops from 900 to 700 div./min., if 5 per cent of coal A from Transvaal is added to coal E, the fluidity decreases by 600 units. On the other hand, dilatometer tests indicated that this mixture gave a better dilatation than the mixture of coal E and 5 per cent anthracite.

Summing .../

Summing up, it may be concluded that as yet no simple test procedure is available giving conclusive evidence of the value of weakly coking coals, especially when these coals are to be blended with coking coals. Consequently, the only way of determining the value of the coals as blend constituents has been to carry out coking experiments, either in experimental coke ovens or under normal full scale operating conditions.

It is envisaged that with the recently constructed 15 inch and 19 inch wide, gas heated experimental coke ovens at the Fuel Research Institute, experimental work will not only be facilitated but that a much wider range of possible blends and operating conditions may be investigated.

D. STUDIES ON METALLURGICAL COKE.

Due to the characteristics of the available coking coals in South Africa the metallurgical industries have to use coke that would probably be regarded as being of low quality or unacceptable for these purposes in Europe or the United States.

The deficiencies are mainly the high ash content, low abrasion index, and to a lesser extent, low shatter index.

For economic reasons the ash in the washed coal cannot be reduced to less than about 10 to 13 per cent, thus giving a coke with an ash content of 15 to 17 per cent.

Various views have been expressed about the probable causes of fissuring in coke[5], and it is generally conceded that fissuring is caused by excessive shrinkage. The opinion has also been expressed that a homogeneous coal, in contrast to a blend, yields a coke with less fissures. This may be true for low or medium volatile coking coals, but experiments conducted with a homogeneous but high volatile (40 per cent D.A.F.) coal from the Transvaal yielded a coke with an abnormally high amount of fissures. It appears that the excessive release of volatile matter is the main cause of the high shrinkage and may overshadow the effect of other factors such as the difference in temperature between the oven wall and centre of the oven and the different rates of shrinkage of the components of a blend.

The poor abrasion index is mainly due to the fact that the weakly and/or non coking particles (consisting mainly of inertinite) in the coal are not sufficiently bonded by the cell-wall material in the coke or the bond may be so thin that its strength becomes less than that of the particles[9] In cokes having an exceptionally low abrasion index many inert particles were found to be only partly bonded. This possibility may be illustrated by reference to a relatively low volatile (25 per cent D.A.F.) coking coal from Natal which was found to contain a very low ratio of active to inert constituents. Microscopical examination of the coke produced from this coal gave evidence of very weak bonds between unfused particles and the cell-wall material surrounding them.

A study of the unfused and/or weakly fused coal particles in 50 samples of coke intermittently manufactured in an industrial coke oven over a period of approximately six months led to the conclusion that the particles of over 3 mm diameter were chiefly responsible for the low abrasion index. A 62 per cent correlation between the occurrence of such particles and the B.S. Cochrane Index was obtained. The correlation between the total amount of unfused particles of over 1 mm diameter and the B.S. Cochrane Index amounted to 55 per cent. No correlation was found for the 3 x 2 mm and 2 x 1 mm particles. The particles of less than 1 mm diameter have probably a beneficial influence on the coke strength. The general view-point that "bone lends strength to the body" has been supported by other authors[5].

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E. SOME RECENT DEVELOPMENTS IN CONNECTION WITH COKE RESEARCH IN SOUTH AFRICA.

In view of the fact that the strength of the coke depends largely on the nature and composition of the coal carbonised and as the choice of raw material is limited, attention is being given to the improvement of the coke by other possible means. The greatest success thus far has been achieved by preheating the coal prior to carbonisation. Preliminary experimental work in this direction yielded results in many respects in agreement with those reported by Perch and Russel[7]. The coal charge was dried by means of superheated steam to 120-180°C, the operation taking from 3 to 5 hours.

Carbonisation was carried out in an electrically heated experimental oven with a capacity of 540 lb. at a bulk density of 50 lb/ft^3.

Two series of tests were carried out. Each series consisted of three tests viz.: (1) a blend charged with a normal moisture content of ca. 7 per cent, (2) the same blend dried at 120°C and (3) the blend dried and preheated to 180°C.

The coal used for the first test series consisted of a mixture of Natal coking coal and three Transvaal weakly coking coals in certain proportions. This mixture is known to give, according to South African standards a reasonably good coke when carbonised under currently accepted conditions. In the second test series the mixture carbonised consisted of a weakly coking coal and a coking coal in a proportion yielding a very poor product under normal carbonisation conditions.

The bulk densities of the moist coal were 47.3 and 42.7 lb/ft^3 for series 1 and 2, respectively. Those of the preheated coals were 54.9 and 49.5 lb/ft^3, respectively (see Table 3).

The physical testing of the coke revealed a considerable improvement in the B.S. abrasion index of the coke manufactured from the dried and preheated coal of both test series. The B.S. shatter indices (on the smaller screens had also been improved but not as extensively as the abrasion index. The relative figures are shown in Table 3.

TABLE 3.

B.S.Shatter and Abrasion Results Pertaining to Cokes from Moist, Dried and Preheated Coal.

Description of Test	Series 1.			Series 2.		
	Moist	Dried at 120°C	Preheated at 180°C	Moist	Dried at 120°C	Preheated at 170°C
Bulk density of coal (dry basis)	47.3	-	54.9	42.7	-	49.5
B.S.Shatter Index:						
+2"	67	64	63	66	70	61
+1½"	83	82	82	83	85	80
+1"	93	94	94	91	94	93
+½"	95.4	97.5	97.5	93.6	96.5	96.4
B.S.Abrasion Index	66	79	79	63	73	73
Bulk density of coke (lb/ft^3 air dry)	26.1	28.5	29.0	27.3	29.3	28.6

Microscopical ..../

Microscopical examination of the various coke samples revealed that the unfused particles in the coke derived from the dried and preheated coals were, in contrast with those samples derived from the moist coal, generally better bonded and in most cases completely surrounded by cell-wall material. This was, presumably, due to the higher bulk density and reduced permeability to escaping volatiles which result in better contact between all particles.

The possibilities of applying a system of selective crushing and screening of the coking coals is also being investigated. Preliminary work[8] has led to the conclusion that South African coking coals, especially the weakly coking coals, are not very amenable to beneficiation by selective crushing and screening and that any benefit accruing from such a process would probably be reflected to only a slight degree in the quality of the coke.

Large scale experimental work to establish the effect of selective crushing, carried out on a weakly coking coal from Transvaal and a Natal coking coal has thus far not yielded very encouraging results. A comparison between the physical characteristics of the coke derived from a mixture of selectively crushed and screened coals and a coke derived from the same mixture but normally prepared prior to carbonisation indicates that no substantial improvement in quality occurred. These results are recorded in Table 4.

TABLE 4.

B.S.Shatter and Abrasion Results Pertaining to Cokes Manufactured from Coal Crushed and Screened in Stages.

Description of Test	Test 1. 70% of A+30% of D normal crushing	Test 2. As for Test 1 but A crushed to -1½mm	Test 3. As for Test 1 but A crushed to -3mm
Bulk density of coal (dry basis)	48.0	43.9	45.7
B.S.Shatter Index:-			
+2"	66	60	62
+1½"	82	78	79
+1"	90	89	90
+½"	92.9	92.0	93.0
B.S.Abrasion Index	61	60	61
Bulk density of coke (lb/ft^3, air dry)	28.6	28.8	28.3

It can be noted from this table that the bulk density of the coal prepared in the normal way was higher than that of the selectively crushed and screened coal. It is also significant that the physical characteristics of the coke prepared from the dried and preheated coals are substantially better than those prepared from the selectively crushed and screened coals.

It would appear that the bulk density of the coal charge is of prime importance in the manufacture of coke, at least from the types of coking coals available in South Africa. Whatever the mechanism may be whereby improved coke results when the charge bulk density is increased, it appears that the advantage achieved in this manner may outweigh those of other refinements available in the preparation of coking charges.

BIBLIOGRAPHY.

1. International Committee for Coal Petrology - Nomenclature Sub-Commission. 1957 Edition.

2. I.S.O./T.C. 27(G.S.-14): Draft recommendation 238, Audibert-Arnu Dilatometer Test for Coal.

3. Hoffmann, H.: Bestimmung der Bildsamkeit nach der Dilatometermethode. - Oel und Kohle in Gem.mit Brennstoff-Chemie Nr 31/32, 15 Aug. 1944.

4. A.S.T.M.Standards on Coal and Coke, Aug. 1947.: Proposed method of test for plastic properties of coal by the Gieseler type plastometer.

5. Mackowsky, M.Th.: Ergebnisse der Koksmikroskopie mit Hilfe der Verschiedensten Untersuchungsmethoden - Brennstoff-Chemie, Nr. 19/20, Bd.36, 12/10/55.

6. Boyer, A.F. : Die mechanischen Vorgänge bei der Verkokung - Brennstoff-Chemie, Nr. 15/16, Bd. 37, 15/8/56.

7. Perch, M.and Russel, C.C. : Preheating coal for carbonisation - Blast Furnace and Steel Plant, 47, 591, June, 1959.

8. Moodie, B. and la Grange, C.C. : Effects of controlled crushing and screening on the analysis and petrographic composition of Blesbok coal. Fuel Research Institute of S.A. Technical Memorandum No.5/1958.
