

# The texture and strength of metallurgical coke

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Attempts to relate the tensile strength of pilot-oven cokes, of blast-furnace quality, to their textural compositions, i.e. the proportions present of the various textural components identifiable in polished coke surfaces under polarized light are described. Of the various equations used, a relationship obtained by multi-linear regression (MLR) analysis permitted the strengths of the cokes from 44 blended coal charges to be calculated with the highest precision but gave no insight into coke breakage behaviour. Equations were therefore derived from consideration of a simple structural model of coke failing in tension by intergranular and transgranular mechanisms. Although predicting coke strengths from textural data with lower precision than the MLR equation, these equations permitted the ready identification of those textural components associated with high strength. Transgranular fracture was indicated as the more probable failure mechanism. A small improvement in precision was obtained by taking the porosity of the coke into account.

## 1. Introduction

The tensile strengths of blast-furnace cokes have been related to coke pore structural parameters determined using computerized image analysis techniques [1]. Of the relationships developed, that considered to be the most soundly based theoretically, i.e.

$$S = KF_{\max}^{-0.5} \exp[-2(F_{\max}/F_{\min})^{0.5} p] \quad (1)$$

where  $K$  is a constant,  $F_{\max}$  and  $F_{\min}$  are maximum and minimum Ferets diameters of the larger pores and  $p$  is the fractional volume porosity, was based on the equation

$$S = KG^{-0.5} \exp(-bp) \quad (2)$$

where  $G$  is the grain size and  $b$  is a constant. This equation was originally developed by Knudsen [2] for ceramics and has been applied successfully by Knibbs [3] to graphites.

Equation 1 was derived without regard to any variation in the nature of the carbon matrix. However, when viewed microscopically, whether using a scanning-electron microscope (SEM) or a polarized-light optical microscope (PLM), coke carbon surfaces appear to be composed of structural units varying in size and shape depending on the rank of the coal carbonized [4, 5]. These induce a characteristic texture to coke surfaces. The three-dimensional nature of these textural components is revealed most readily by SEM examination of fractured coke surfaces [4]. Differences thus evident in the surface topography of the various components imply variations in their mode of failure and consequently in their contribution to coke strength. Any such effect would be dependent upon the coke textural composition, i.e. the proportion of the various components present. In preliminary studies, subjecting tensile strength and SEM textural data to multi-linear regression analysis [6] showed that

it was possible to relate strength and textural data. Coke textural components can be classified in such a way that broad correspondence between data obtained using SEM and PLM methods of assessment is achieved [7]. The present work describes attempts to relate coke strengths and PLM textural data. In addition to applying multilinear regression analysis, relationships derived from consideration of failure of a simple coke structural model were also investigated.

## 2. Experimental procedure

### 2.1. Cokes used

The cokes used were produced by carbonizing 44 multicomponent coal charges, sized 90 wt % less than 3 mm, in a small pilot oven [8] under conditions chosen to give dense, high-strength coke. The tensile strengths of the cokes were determined by the diametral-compression method [9] using an Instron universal testing machine, a cross-head speed of  $0.5 \text{ mm min}^{-1}$  being employed. The mean tensile strengths quoted are average values obtained using at least 30 specimens, 10 mm long by 10 mm diameter.

### 2.2. Determination of PLM textural data

To prepare coke samples for examination under polarized light, they were first crushed gently to maximize the yield of material in the 120 to  $600 \mu\text{m}$  size range. After removal of fines, by ultrasonic cleaning, and drying, the coke grains were mixed with epoxy resin and formed into a 15 mm diameter pellet. This was then embedded into further resin and cured to form a 25 mm diameter by 10 mm thick block. The upper, coke-bearing surface was then polished, using conventional methods, to give a scratch-free, highly reflecting surface.

PLM textural data were then determined using a Leitz Ortholux polarizing microscope. Crossed polars,

together with a full wave retarder plate, were used to impart colour to the image and a  $\times 100$  air objective and  $\times 10$  eye-pieces were used to give an overall magnification of  $\times 1000$ . Textural data quoted are based on the examination of 1000 positions on the coke surface of each block. A Swift mechanical stage and electronic counter were used to position the block and to accumulate the data. At each position, the textural component present under the cross-wires was allocated to one of the nine textural categories described in Table I. Detailed descriptions, with colour micrographs, of the various components have been given previously [7]. Briefly, the components of particular interest fall into broad classes termed flow and mosaic. Both classes are divided into three sub-classes in order adequately to differentiate between cokes. Flow components are evident as assemblies of elongated isochromatic areas while mosaics consist of groups of rounded isochromatic units. Both originate from the coal maceral vitrinite which softens during carbonization. Also present in the cokes are isotropic vitrinite-derived material and isotropic carbonaceous inert particles. The sizes quoted in the table for the various mosaics are mean values obtained from a total of 300 measurements taken from projected images of six transparencies, each showing material typical of the component considered. From each transparency, measurements were taken from isochromatic areas believed to represent the basic unit constituting the mosaic.

### 3. Results

Table II lists the measured fractional textural compositions of the 44 cokes, along with their tensile strengths. It is assumed that the densities of the various components are equal so that although data were measured as fractions vol/vol, they can also be regarded as fractions wt/wt. Cokes are arranged in order of increasing tensile strengths which range from 4.42 to 6.96 MPa, such values being comparable to those of good-quality blast furnace cokes. All cokes contained both large and small inert components, the

fractional contents ranging from 0.012 to 0.186 and 0.056 to 0.107, respectively. The other textural components, listed in the table, were present in the cokes in proportions related to the ranks of the coals in the blend, up to a maximum fractional value for any single component of 0.540.

### 4. Discussion

Applying multi-linear regression analysis to the tensile strength and textural composition data resulted in the following equation

$$S = 0.454 + 4.37Fb - 1.71Fs + 6.96Fg + 4.84Mc + 4.80Mm + xxMf + 2.65I + 3.8Inl + 14.91Ins \quad (3)$$

where the initials refer to the fractional content of the textural component identified by the initials used in Table I. The statistical package used eliminated fine mosaics (Mf) from consideration on the grounds that the textural contents correlated highly with those of other components. Strength values calculated from this equation are compared with measured values in Table III. The equation permits the strengths of the cokes to be calculated with a standard error of estimation of 0.41 MPa. This is approximately twice the error associated with the tensile strength measurement. However, a purely statistical relationship of this type is of little scientific merit. It gives neither insight into coke breakage nor ready identification of those textural components associated with high coke strength.

SEM fractographic studies [4] indicated that flow components were composed of lamellae predominantly aligned circumferentially to pore surfaces. Crack propagation from pore to pore thus results in trans-lamellar fracture. In contrast, mosaic components appeared to fail by an intergranular mechanism. Accordingly further attempts to relate coke strength and textural data were made using equations derived from consideration of a simple model of coke failing in tension by transgranular and intergranular

TABLE I PLM textural component classification

Component	Initial	Appearance of polished surface
Anthracitic:		A non-porous anisotropic component of cokes made from high-rank coals, which does not merge with mosaic components.
plain	A	Single-coloured particles.
patterned	Ap	Particles with layered structure of contrasting colour.
Flow:		Composed of elongated isochromatic areas often curved round pores.
broad	Fb	Size $> 20 \mu\text{m} \times > 10 \mu\text{m}$ .
striated	Fs	Size $> 20 \mu\text{m} \times > 2 \mu\text{m}$ .
granular	Fg	Size $> 3 \mu\text{m} \times > 1 \mu\text{m}$ .
Mosaic:		Composed of small rounded isochromatic areas.
coarse	Mc	Mean size $0.91 \mu\text{m}$ .
medium	Mm	Mean size $0.63 \mu\text{m}$ .
fine	Mf	Mean size $0.50 \mu\text{m}$ .
Isotropic:	I	An optically isotropic component of cokes from low-rank coals. Fuses well to mosaic components.
Inerts:	In	Carbonaceous inerts are isotropic components identifiable by their woody structure or, if small, by their unfused sharp edges. Mineral matter is included in this class.
large	Inl	Size $> 50 \mu\text{m}$ .
small	Ins	Size $< 50 \mu\text{m}$ .

mechanisms. Coke is assumed to be composed of a regular array of close-packed, equi-sized cubic grains and that the nine textural components are randomly distributed within each layer. Intergranular failure is then simply regarded as the pulling apart of two such layers along interfaces between textural components while in transgranular breakage the fracture path passes through the components constituting the layer. The simplicity of this approach is acknowledged. Because no account is taken of any flaws present, the approach can be reconciled with a Griffiths view of brittle fracture only if it is assumed that the flaws are a constant factor.

For intergranular fracture, coke strength is dependent on the probability of contact of grains, of the various textural types, across the interface between two layers and the strength of the bond between them. Then the strength,  $S$ , is given by

$$S = \sum_{i=1}^9 \sum_{k=1}^9 F_i F_k S_{i,k} \quad (4)$$

where  $i$  may equal  $k$  and  $F_i$  and  $F_k$  are fractional textural contents of the  $i$ th and  $k$ th components and  $S_{i,k}$  is the intercomponent strength. When  $i = k$  then the strength term can be regarded as the strength of a coke composed of a single component failing by an intergranular mechanism.

In the case of transgranular fracture, coke strength is dependent upon the probability of occurrence of the various textural components in a layer and their strength. Then

$$S = \sum_{i=1}^8 F_i S_i \quad (5)$$

where  $F_i$  has the same meaning as before and  $S_i$  is the transgranular failure strength of a single-component coke.

These equations were fitted to the data using a reiterative computer method in which strength values, giving the lowest standard error of estimation, were sought from a preselected limited number of values for each textural component. Running the programme

TABLE II PLM textural composition of cokes

Coke number	Tensile strength (MPa)	Fractional textural composition								
		I	Mf	Mm	Mc	Fg	Fs	Fb	Inl	Ins
1	4.42	0.055	0.195	0.345	0.035	0.126	0.045	0.014	0.012	0.065
2	4.43	0.051	0.233	0.318	0.028	0.084	0.042	0.026	0.129	0.089
3	4.52	0.074	0.170	0.157	0.028	0.176	0.106	0.039	0.151	0.099
4	4.52	0.018	0.037	0.232	0.068	0.192	0.113	0.178	0.093	0.069
5	4.65	0.036	0.134	0.204	0.027	0.215	0.117	0.055	0.142	0.070
6	4.90	0.031	0.093	0.132	0.023	0.242	0.164	0.106	0.131	0.078
7	4.91	0.018	0.052	0.325	0.071	0.164	0.085	0.144	0.067	0.074
8	5.02	0.012	0.002	0.043	0.238	0.368	0.108	0.070	0.095	0.064
9	5.09	0.030	0.187	0.389	0.052	0.096	0.030	0.033	0.107	0.076
10	5.11	0.014	0.036	0.377	0.168	0.139	0.048	0.026	0.132	0.060
11	5.17	0.006	0.007	0.144	0.190	0.307	0.122	0.080	0.107	0.067
12	5.20	0.029	0.116	0.229	0.044	0.264	0.072	0.022	0.135	0.089
13	5.25	0.010	0.006	0.031	0.194	0.392	0.125	0.070	0.115	0.057
14	5.27	0.054	0.135	0.352	0.036	0.197	0.039	0.007	0.105	0.075
15	5.30	0.009	0.007	0.076	0.068	0.441	0.095	0.090	0.135	0.075
16	5.31	0.034	0.058	0.295	0.060	0.128	0.084	0.149	0.114	0.078
17	5.32	0.024	0.156	0.278	0.052	0.218	0.032	0.100	0.084	0.056
18	5.35	0.023	0.074	0.478	0.061	0.057	0.036	0.050	0.114	0.107
19	5.40	0.004	0.029	0.209	0.149	0.238	0.084	0.078	0.135	0.074
20	5.41	0.028	0.169	0.330	0.057	0.155	0.044	0.055	0.084	0.078
21	5.43	0.008	0.034	0.291	0.159	0.211	0.075	0.046	0.101	0.075
22	5.51	0.001	0.022	0.519	0.137	0.095	0.032	0.029	0.104	0.061
23	5.59	0.017	0.042	0.113	0.026	0.424	0.122	0.044	0.136	0.076
24	5.69	0.014	0.037	0.098	0.037	0.384	0.159	0.052	0.133	0.086
25	5.69	0.036	0.114	0.170	0.060	0.204	0.109	0.119	0.128	0.060
26	5.69	0.004	0.010	0.172	0.287	0.215	0.067	0.039	0.109	0.097
27	5.70	0.021	0.043	0.400	0.048	0.095	0.075	0.116	0.110	0.092
28	5.76	0.010	0.003	0.015	0.024	0.450	0.225	0.064	0.130	0.079
29	5.77	0.014	0.077	0.026	0.020	0.453	0.178	0.026	0.186	0.090
30	5.79	0.012	0.032	0.189	0.028	0.172	0.125	0.217	0.133	0.092
31	5.87	0.058	0.112	0.158	0.050	0.317	0.063	0.014	0.150	0.078
32	5.94	0.020	0.042	0.406	0.081	0.107	0.063	0.075	0.114	0.092
33	6.05	0.011	0.041	0.460	0.108	0.192	0.013	0.005	0.084	0.086
34	6.06	0.026	0.025	0.203	0.092	0.336	0.058	0.074	0.091	0.095
35	6.10	0.016	0.053	0.541	0.081	0.093	0.031	0.034	0.079	0.072
36	6.15	0.014	0.047	0.372	0.085	0.230	0.036	0.035	0.111	0.070
37	6.16	0.011	0.004	0.015	0.053	0.447	0.123	0.102	0.161	0.084
38	6.27	0.001	0.012	0.141	0.216	0.284	0.059	0.075	0.139	0.073
39	6.28	0.025	0.016	0.148	0.077	0.439	0.041	0.035	0.122	0.097
40	6.29	0.023	0.063	0.503	0.058	0.124	0.023	0.020	0.084	0.102
41	6.64	0.015	0.011	0.032	0.069	0.417	0.117	0.083	0.166	0.100
42	6.64	0.010	0.010	0.188	0.290	0.252	0.069	0.052	0.062	0.067
43	6.78	0.010	0.003	0.028	0.260	0.408	0.077	0.031	0.115	0.068
44	6.96	0.012	0.001	0.061	0.246	0.400	0.067	0.046	0.097	0.070

several times produced a good, but not necessarily the best, fit.

The values of the strength terms obtained by fitting Equation 4 to the data are listed in Table IV at the intersections of the rows and columns. Those values lying along the diagonal are the strengths of the interface between two grains of the same component type while the other values are intercomponent strengths. The highest values, lying within the odd-shaped area, indicate those textural components, i.e. granular flow and medium and coarse mosaics, associated with high coke strength. The standard error of estimating the coke strengths using this equation and the strength values listed is 0.49 MPa.

Applying Equation 5, i.e. that derived on the basis of transgranular fracture, gave the following relationship

$$S = 1.9I + 2.5Mf + 6.5Mm + 5.9Mc + 8.2Fg + 2.5Fs + 4.4Fb + 4.1In \quad (6)$$

where the initials again refer to the fractional content of the identified textural component. In fitting this equation the inert components, irrespective of size, were considered as a single category. This relationship permits the coke tensile strengths to be calculated with a standard error of estimation of 0.46 MPa. This represents a small improvement over that achieved using Equation 4, so that transgranular fracture is indicated as the more probable mode of coke breakage.

Although the relationships based on Equations 4 and 5 do not permit the calculation of coke strength with a precision equal to that of the multi-linear regression relationship (Equation 3) they do permit the ready identification of those textural components associated with high coke strength. Both indicate that high contents of granular-flow and medium and coarse mosaics are required. Being simplest in form, Equation 6 appears most suitable for this purpose.

It has previously been considered that mosaic units in coke carbon consist of compressed mesophase units [10]. However, during this work, careful examination of textural components, under polarized light at high magnification, showed that the shape of the isochromatic areas in all optically anisotropic textural components change on rotation of the specimen stage. This implies that they all consist of continuous lamellar structures, the size of the isochromatic areas reflecting that of volumes of commonly aligned lamellae. On this basis, fracture will involve trans-lamellar breakage or interlamellar cleavage. It is now considered that it is the latter effect, occurring in mosaic components, which gives the impression of intergranular failure when fracture surfaces are viewed in an SEM.

No detailed pore structural data were accumulated for the cokes made in this study, but coke porosities, available from density measurement were available. These values, listed in Table III, were used in an attempt to modify the equation, derived from transgranular failure considerations, to reflect variations in the porosities of the cokes. The equation used is

$$S = \sum_{i=1}^9 S_i T_i \exp(-bp) \quad (7)$$

TABLE III Comparison of measured and calculated coke tensile strengths

Coke number	Measured values		Coke tensile strengths calculated using Equation			
	Tensile strength (MPa)	Fractional porosity	3	4	6	9
1	4.42	0.579	4.71	5.00	5.01	4.87
2	4.43	0.589	4.70	4.63	4.71	4.38
3	4.52	0.544	4.80	4.71	4.66	4.75
4	4.52	0.559	5.25	5.38	5.34	5.12
5	4.65	0.555	4.78	5.10	5.06	5.07
6	4.90	0.543	4.81	5.10	5.00	5.03
7	4.91	0.560	5.39	5.58	5.46	5.40
8	5.02	0.541	5.84	6.11	5.96	5.97
9	5.09	0.600	4.95	5.04	5.12	4.70
10	5.11	0.584	5.51	5.70	5.72	5.50
11	5.17	0.538	5.62	5.87	5.78	5.91
12	5.20	0.567	5.49	5.40	5.45	5.42
13	5.25	0.546	5.68	6.05	5.92	5.87
14	5.27	0.583	5.31	5.45	5.42	5.29
15	5.30	0.544	6.10	5.93	6.04	6.20
16	5.31	0.549	5.24	5.17	5.18	5.22
17	5.32	0.553	5.16	5.38	5.43	5.47
18	5.35	0.583	5.68	5.33	5.38	5.30
19	5.40	0.546	5.66	5.59	5.68	5.80
20	5.41	0.574	5.11	5.25	5.24	5.09
21	5.43	0.550	5.68	5.84	5.77	5.99
22	5.51	0.593	5.65	6.00	5.90	5.70
23	5.59	0.551	5.75	5.92	5.87	6.03
24	5.69	0.547	5.56	5.76	5.65	5.80
25	5.69	0.528	4.79	5.06	5.05	5.27
26	5.69	0.557	6.09	5.73	5.79	5.65
27	5.70	0.563	5.49	5.36	5.84	5.38
28	5.76	0.563	5.37	5.92	5.66	5.47
29	5.77	0.565	5.72	5.69	5.74	5.62
30	5.79	0.563	5.34	4.95	5.10	5.21
31	5.87	0.561	5.50	5.42	5.96	5.47
32	5.94	0.570	5.62	5.48	5.47	5.46
33	6.05	0.575	6.15	6.11	6.08	6.18
34	6.06	0.555	6.26	5.96	6.26	6.04
35	6.10	0.576	5.60	5.93	5.77	5.88
36	6.15	0.576	5.84	5.91	5.94	5.90
37	6.16	0.563	6.02	5.69	5.87	5.60
38	6.27	0.535	6.00	5.74	5.90	6.11
39	6.28	0.572	6.65	6.13	6.26	6.10
40	6.29	0.579	5.96	5.78	5.74	5.81
41	6.64	0.560	6.17	5.60	5.81	5.64
42	6.64	0.548	5.88	6.16	5.97	5.99
43	6.78	0.551	6.17	6.27	6.16	6.08
44	6.96	0.546	6.25	6.23	6.21	6.22

and is based on the Ryshkewitch-Duckworth equation:

$$S = S_0 \exp(-bp) \quad (8)$$

where  $p$  is the fractional porosity and  $b$  is a constant unless pore shape varies. In Equation 8,  $S_0$  is considered to be the strength of a non-porous body. On

TABLE IV Strength terms obtained by fitting Equation 4 to the experimental data

	Inl	Ins	Fb	Fs	Fg	Mc	Mm	Mf	I
I	4.0	5.5	3.4	3.0	5.2	5.2	5.2	3.0	3.4
Mf	4.0	5.5	3.0	3.0	6.0	4.4	3.0	3.0	
Mm	4.1	5.0	5.2	7.0	7.5	6.5	7.5		
Mc	4.1	5.0	5.2	3.5	8.3	5.5			
Fg	4.1	5.0	5.2	7.5	9.1				
Fs	4.0	5.5	3.0	3.5					
Fb	4.0	5.5	3.4						
Ins	0.0	0.0							
Inl	0.0								

this basis, the summation term in Equation 7 can be regarded as indicating the varying contribution of the textural components to the strength of pore-free cokes.

Studies of the porous structure of cokes, using automated image analysis [1], showed that, for most blast-furnace cokes, the mean aspect ratio of pores in cokes,  $F_{\max}/F_{\min}$ , lay within the range 1.72 to 1.90. Using the expression for  $b$  in Equation 1, these values correspond to values for  $b$  of 2.62 to 2.76. An explicit derivation of the Knudsen relationship has since been published [11]. This included, from consideration of far-field displacement effects, the derivation of an alternative expression for the variation of  $b$  with pore shape. On this basis, for aspect ratios of 1.0 to 2.0,  $b$  remains almost constant at 2.78. Therefore, in attempting to relate coke strength and textural data by evaluating the strength terms in Equation 7, only a  $b$  value of 2.8 was used. The following relationship was obtained

$$S = (8I + 8Mf + 36.5Mm + 23.5Mc + 40.5Fg + 10.5Fs + 11Fb + 19In) \exp(-2.8p) \quad (9)$$

From this equation, the coke tensile strengths can be calculated with a standard error of estimation of 0.45 MPa so that only a small improvement in precision had resulted from the incorporation of a porosity term. The strengths calculated using this equation are compared with measured values in Table III.

The standard errors of estimation obtained in this work imply that the equations account for approximately 70% of the variation in tensile strength of the cokes. However, whether the tensile strength of coke is causally dependent upon coke textural composition to the extent implied by the low standard errors of estimation is open to question. It is recognized that for coal blends carbonized under a single set of carbonizing conditions, changes in blend composition lead

simultaneously to changes in both pore structural characteristics and textural composition. Thus, it is difficult to obtain a relationship between coke tensile strength and either pore structural parameters or textural data individually which excludes an effect due to the other factor. Hence individual approaches can both account for a high proportion of the variation in tensile strength. Thus, although on theoretical grounds, coke strength is expected to be dependent upon both the porous structure and the nature of the coke matrix, only after further careful work will the relative contributions of the porous structure and textural composition be separately identified.

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