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An SEM study of the tensile fracture of metallurgical coke

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An SEM study of the tensile fracture of an experimental blast-furnace coke is described, the objective being to further the understanding of the failure of coke under diametral compressive loading and the influence thereupon of the textural components of the coke carbon. The approach adopted was to examine etched surfaces of the central spherical plane of cylindrical test specimens after loading diametrically. For each specimen, a diagram was drawn indicating the number, size and position of the flaws present, a note being made of the textural component through which the flaw passed. It was found that textural components were present at flaws in the same proportions as in the coke as a whole, implying that the initiation and propagation of microcracks is not influenced by any variation in the properties of the coke textural components. Fracture resulted when microcracks, initiated at pores in the coke at lower stress levels, joined together to form a flaw of critical size. Fracture crack systems were very complex, but nevertheless always contained a diametral fracture, this being indicative of a valid test.

1. Introduction

Metallurgical coke for blast-furnace use is produced by the carbonization of blended coal charges in slot-type ovens. Such coke, in common with most other carbons, exhibits brittle behaviour [1], fracture being governed by the Griffiths criterion and the strength by the size of the largest flaws present [2]. The tensile strengths of cokes, determined by the diametral compression method, have been related to parameters describing their porous structure, the form of the equation used implying that the larger pores act as the Griffiths critical flaws [2]. In this work, no account was taken of any variation in the nature of the carbon in the coke matrix although the coke carbon is composed of microscopically identifiable textural units, the size and shape of which vary depending on the rank of the coal carbonized [3]. The three-dimensional form of these units can be directly observed when coke fracture surfaces are viewed in a scanning electron microscope. On the basis of their appearance in both fractured and etched surfaces, the units have been classified into broad categories termed flat, lamellar, intermediate, granular and inert [3], but in order to differentiate adequately between cokes from low-rank coals it was necessary to sub-divide the granular category into four depending upon the size of the component "grains".

Variations in the roughness of the fracture surfaces of the various textural components and the differences in the mode of fracture thus evident imply variations in their strength. Accordingly, an attempt has been made to study the initiation and propagation of cracks in coke, the objective being to further the understanding of coke breakage in the diametral compression test and to assess the influence thereupon of the various coke textural components. The approach adopted was to compare, by examination in a scanning electron microscope, coke specimens before and after stressing by diametral compressive loading.

2. Experimental procedure

2.1. Coke used

The coke studied was produced in a 17-tonne test oven [4] by carbonizing a wet-charged blend containing a 2 : 3 : 1 : 4 by weight mixture of coals in International Classes 334, 434, 635 and 634. It was anticipated on the basis of the known variation of textural composition with the rank of coal carbonized [3] that the coke would contain appreciable quantities of lamellar, intermediate and medium and fine granular carbon, together with inert components which had not fused during carbonization. Micrographs illustrating the various textural types have been published previously [3]. They are described in Table I.

The tensile strength of the coke was determined by the diametral compression method [1] using a Tensometer universal testing machine, a crosshead speed of 0.5 mm min^{-1} being employed. The mean strength value was obtained using forty-nine 10 mm diameter by 10 mm long cylindrical specimens, the mean value obtained, 5.1 MPa, being comparable to that for good-quality blast furnace coke.

2.2. Specimen preparation and examination

The following four types of specimen were examined in the SEM:

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TABLE I Appearance of textural components

Component type	Appearance of etched surface
Flat (F)	Generally rather flat, sometimes with a fine granularity. Some regions contain scattered circular pits or short, narrow channels.
Lamellar (L)	Surface consists of parallel ridges and channels $> 5\,\mu$m long.
Intermediate (I)	Intermediate in appearance between lamellar and granular forms with short ($< 4\,\mu$m) channels, often branched.
Granular (G):	Uniform, pitted texture.
Coarse (Gc)	Pit size approximately 0.2 to $0.35\,\mu$m.
Medium (Gm)	Pit size approximately 0.15 to $0.2\,\mu$m.
Fine (Gf)	Pit size approximately 0.1 to $0.15\,\mu$m.
Very fine (Gcf)	Pit size approximately $< 0.01\,\mu$m.
Inerts (Ins)	Identifiable by their woody structure or, if small, by their unfused sharp edges. Particles often appear darker and more deeply etched than the reactive matrix.

(a) "as-received" specimens;

(b) "stressed" specimens, which had survived a load equivalent to the mean tensile strength of the coke;

(c) "stress-relieved" specimens, the loading of which was discontinued when a marked fall in the force–displacement curve indicated marked stress relief;

(d) "fractured" specimens, i.e. after loading to failure.

It should be noted that as regards their strength the as-received, stress-relieved and fractured specimens should be more easily comparable with the coke sample as a whole than the stressed specimens, all of which belonged to the stronger half of the sample. It is calculated from the dispersion of strength values that the latter specimens had been stressed to within 50 to 100% of their breakage stress.

To prepare samples for examination in the SEM, ten specimens of each type were first embedded in epoxy resin, leaving 5 mm of their length standing proud. After curing the resin, the exposed coke was abraded away and the surface of the cross-sectional plane at the centre of the specimen polished by standard techniques. Polished surfaces were then etched in atomic oxygen formed in an electrodeless discharge and gold-coated prior to examination in the SEM. At all stages of preparation and examination care was taken to ensure that the orientation of the stressed diameter was known to within about $\pm 10°$.

These specimens were examined in the SEM using fourteen equally spaced traverses at an instrument magnification of × 200.

Under these conditions the whole of the surface could be viewed with minimum overlap. The positions along the traverse of any small flaws were noted, as were the textural components through which they passed. For extended microcracks and fracture cracks (the terms are explained fully below) this procedure was repeated for each traverse in which the crack was observed, the textural component at two positions within each field of view being noted.

The textural composition of the coke was determined as described previously [3]. Briefly, a point-counting technique was applied during the SEM examination of etched surfaces of uncrushed coke. Data quoted are based on 500 positions on the coke surface, the textural component present at each position being allocated to one of the textural categories in Table I.

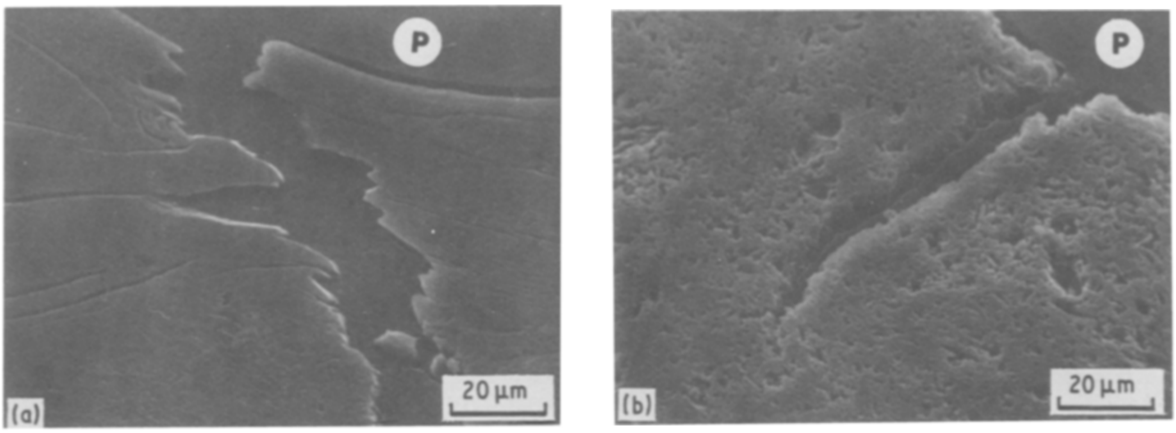
3. Results

Preliminary examination of specimens in the SEM showed that the flaws present in the samples examined varied from microcracks, which extended from a pore into or across the adjoining pore wall, to extended microcracks, larger fissures at least 300 μm in size and evident on more than one traverse, and ultimately to major fracture cracks traversing the whole specimen. Accordingly, when mapping their distributions, flaws were classified into one of these three categories. Interlamellar fissures in lamellar carbon were discounted unless they originated at a pore. Also, minor flaws near the edges of specimens which were considered to have arisen during specimen preparation were ignored.

The appearance of the three types of microcrack is illustrated in the micrographs in Figs 1 to 3. In lamellar coke carbon, the lamellae are usually aligned circumferentially to the pore surface so that crack propagation from pore to pore results in breakage across the lamellae as shown in Fig. 1a. The jagged nature of the crack path is attributed to the propagating crack temporarily being diverted along relatively easy propagation pathways between lamellae. Fissures in intermediate and medium granular components are shown in Figs 1b and c. The variation in tortuosity of the cracks shown is commensurate with the acknowledged variation in the size and shape of the textural components present.

The carbonaceous inert components in the coke were invariably found to be enfolded within reactive-derived components. For present purposes flaws were regarded as being associated with inert components if they extended from a pore and traversed reactive-derived constituents to the inert particle (Fig. 2). More extensive cracks are illustrated in Fig. 3, a network of extended microcracks being shown in Fig. 3a and part of a fracture crack in Fig. 3b.

Flaw distribution diagrams obtained using the four types of specimen are shown in Figs 4 to 7, the orientation of the stressed diameter being from top to bottom as indicated. The values beneath each individual flaw diagram in Figs 4 to 6 refer to the number of flaws observed in that specimen. Within each group of specimens considerable variation in the number of flaws per specimen was observed. However, it is clear that the stress-relieved and stressed specimens contained a larger number of both simple and extended microcracks than the as-received specimens. The average number of simple microcracks per specimen for the as-received, stressed and stress-relieved specimens was 37, 47 and 54, respectively. The total number of extended microcracks, indicated in the flaw distribution diagrams by short lines, observed in all



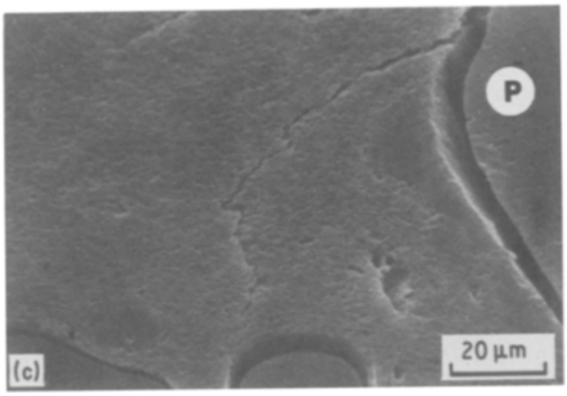


Figure 1 Microcracks running from a pore (P) into a cell wall consisting of (a) lamellar, (b) intermediate and (c) medium granular coke carbon.

ten specimens of the three types was 14, 28 and 34, respectively.

Crack diagrams for the fractured coke specimens are given in Fig. 7, the observed specimen strength being given under each individual crack diagram. The mean tensile strength for these specimens, 4.71 MPa, is slightly lower than that of the sample as a whole, but the spread of results is similar. Positions of microcracks in these specimens were not recorded. The figure demonstrates clearly the complexity of the crack networks resulting from breakage of this coke under diametral compression. Multiplanar and branched cracking is the rule rather than the exception. In one or two instances it is suspected that the orientation of the loaded diameter had not been maintained accurately during sample preparation, but this does not obscure the fact that in certain cases a subsidiary crack had propagated at a high angle to the loaded diameter.

The frequency of observation of the various textural components at microcracks in the as-received, stressed and stress-relieved specimens, and at fracture cracks in the fractured specimens, is expressed as a percentage and compared with the textural composition of the coke in Table II, the textural components being identified by their initial letter as in Table I.

The relatively low number of microcracks associated with carbonaceous inerts stems from their immersion within the cell-wall material. When account is taken of this factor, for the reactive-derived components there is a broad correspondence between the textural composition and the frequency of observation of textural components both at microcracks and at fracture cracks.

4. Discussion

Metallurgical cokes are notoriously inhomogeneous [5]. Thus individual cokes made using a blend of a number of coals of varying rank are likely to contain all the microscopic features evident in all commercial cokes. In the present study, numerical data were therefore accumulated during microscopic observation in an attempt to make valid deductions. However, in view of the relatively small number of specimens examined it is doubtful whether the microcrack density data in particular can be regarded as more than semi-quantitative. Furthermore it is recognised that this study represents an attempt to investigate a three-dimensional effect by examining two-dimensional features.

Variations in the mode of fracture of the various textural components have been inferred previously from SEM studies of fracture surfaces [3]. Additional supporting evidence is now available from the micrographs illustrating crack paths in Figs 1 to 3. The irregularity of the crack path through granular components clearly varies depending on the pit sizes

TABLE II Comparison of textural composition and occurrence of textural components at cracks

Property	Frequency of observation of components (%)							
	Ins	F	L	I	Gc	Gm	Gf	Gvf
Textural composition	12	0	17	31	3	17	15	5
Components at:								
microcracks	2	0	20	36	2	20	15	4
fracture cracks	6	0	19	38	3	16	16	3

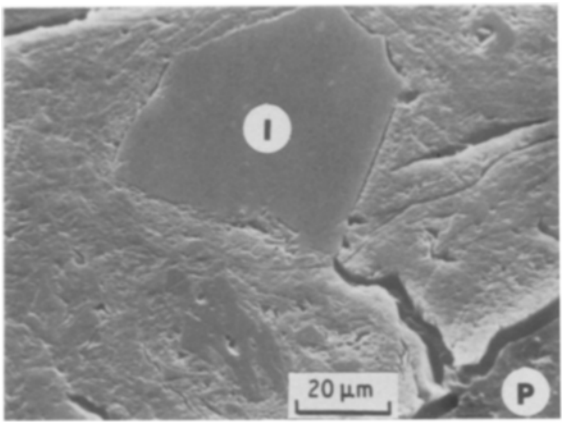


Figure 2 Microcracks running from a pore (P) to an inert particle (I).

observed in the etched surface, thus supporting an intergranular fracture mechanism. Fracture of lamellar components aligned circumferentially to the pore surface involves breakage across the lamellae. The jagged fracture path shown in Fig. 1a is typical of a tough material. It is this difference in the mode of fracture which suggests a corresponding variation in the porosity-free strength of the different coke carbon textural components. For the lamellar and larger granular components the observed large angular diversion of the crack paths may be indicative of shear stresses as well as tensile stresses being involved in crack propagation.

As Table II indicates, the frequency of observation of the various textural components at both fracture cracks and microcracks corresponds to that expected on the basis of the textural composition of the coke. Thus, in contrast to previously expressed views [6], there appears to be no marked preference for cracks to be initiated in or diverted through any particular textural component. It would appear therefore that either crack initiation and propagation is determined primarily by the stress distribution, or stress concentration effects overshadow any variation in the mechanical properties of the textural components.

Regarding the mechanism of coke breakage, with the techniques available it was not possible to observe directly the various stages leading to fracture. Nevertheless, from the data it is possible to construct the following sequence of events.

Since coke specimens subjected to diametral compressive loading contained a higher number of microcracks and extended microcracks than the as-received specimens, the implication is that stable microcracks are introduced into the coke structure by a stress less than the breakage stress. The stresses associated with shrinkage during the later stages of carbonization, thermal shock during quenching, and mechanical shock during handling are considered to be responsible for the microcracks seen in the as-received specimens as well as for the gross fissures visible by eye in lump coke. The variation of the microcrack density in the as-received specimens implies differing degrees of local pre-stressing.

It is not possible to identify microcracks induced by diametral compressive loading. However, the flaw distribution diagrams for the stressed specimens do not suggest any marked tendency for microcracks to be concentrated in the area of high tensile stress along the loaded diameter. The ratio of compressive to tensile stress increased with distance from the centre of the specimen [7], but since the initiation of microcracks also depends on other factors, for example the inclination of the stress-concentrating flaw to the direction of the applied stress, microcracks formed away from the centre of the specimen are not necessarily induced by compressive forces.

Most simple microcracks were observed to extend from a pore into the cell wall or to traverse the cell wall to an adjacent pore. This implies that pores are the principal centres for microcrack initiation. Since so many simple microcracks were observed to extend only from one pore to another, pores appear to have the ability both to initiate and to stabilize microcracks. Stabilization is presumed to stem from the broadened crack tip no longer acting as an effective stress raiser so that crack propagation is interrupted. No obvious difference in the shape of those pores which acted as initiators or stabilizers of microcracks was apparent in the two-dimensional view obtained from etched surfaces. The formation of stable microcracks will be associated with dissipation of strain energy. This effect, together with some edge-crushing near the platens, is responsible for the applied load often increasing in a stepwise manner and for the audible creaks and groans emitted by coke specimens under load.

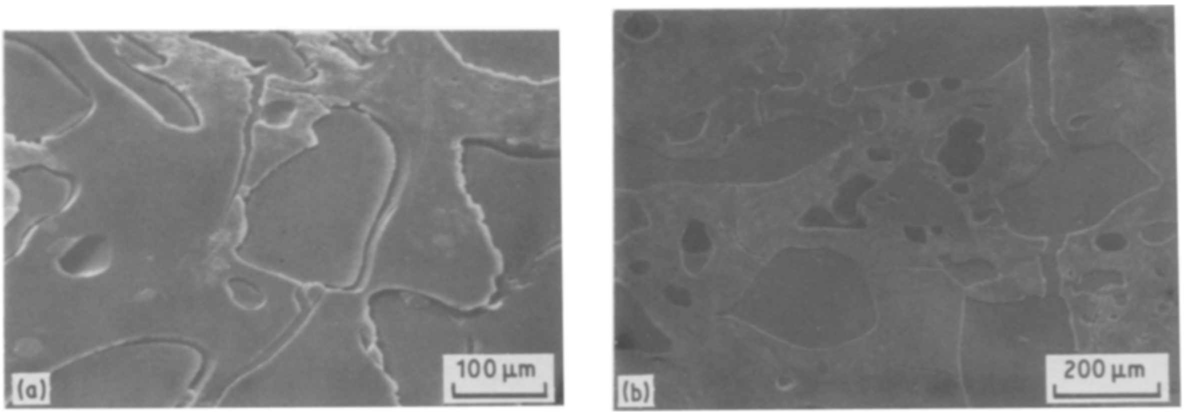


Figure 3 (a) Extended microcrack and (b) fracture crack networks

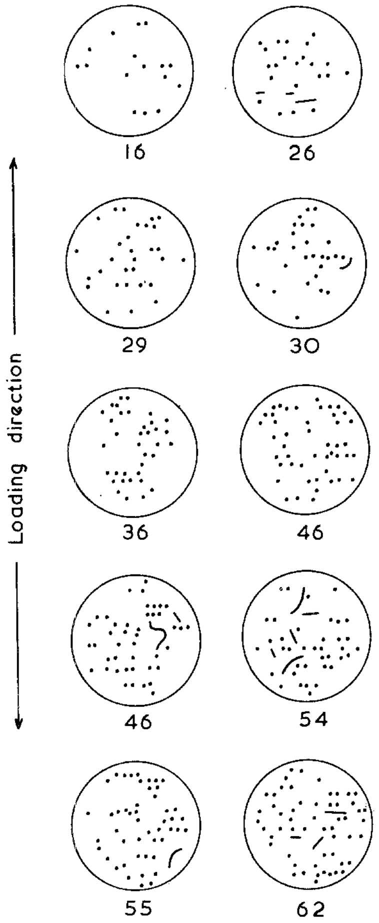


Figure 4 Flaw distribution diagrams for "as-received" specimens. The numbers refer to the number of flaws per specimen.

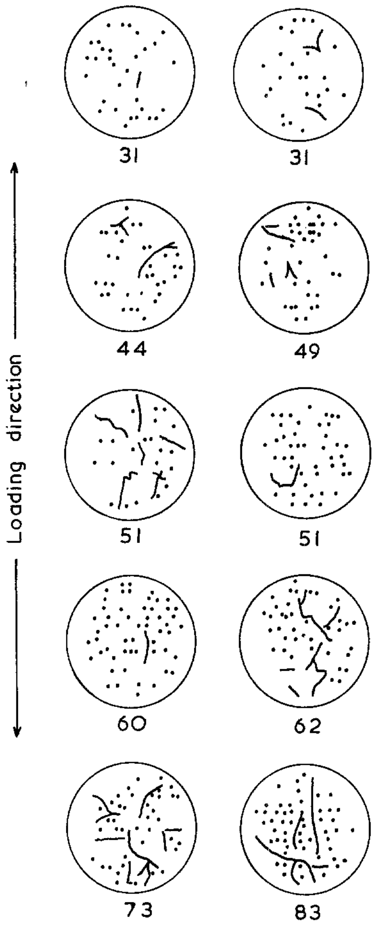


Figure 6 Flaw distribution diagrams for "stress-relieved" specimens.

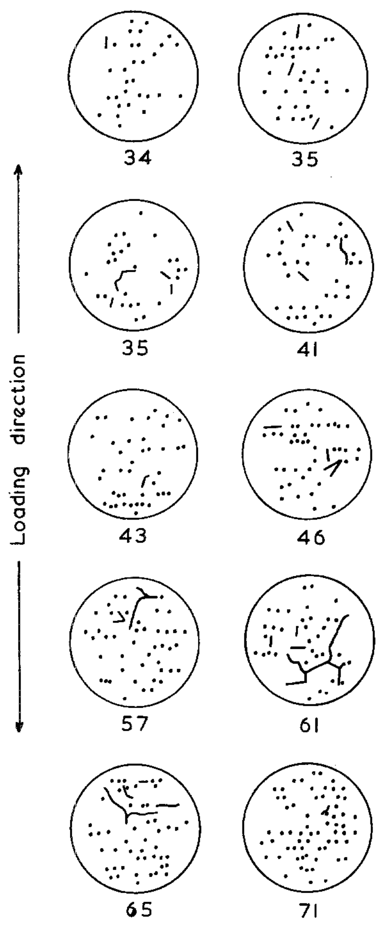


Figure 5 Flaw distribution diagrams for "stressed" specimens.

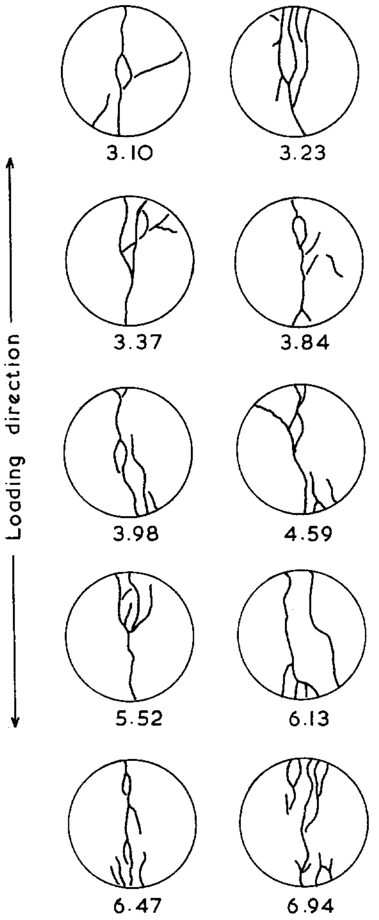


Figure 7 Fracture crack diagrams. The numbers give the observed specimen strength (MPa).

Initially each further increase in the load can be envisaged to initiate a new generation of discrete subcritical microcracks many quite remote from those formed earlier, and these too grow only to limited sizes before being stabilized. At high stress levels, when the microcrack density is already high, further loading of the specimen induces the formation of extended microcracks, these often being associated with a much higher degree of stress relief and consequent fall-back of the applied load. It is evident from Fig. 6 that the length of stable extended microcracks can approach 5 mm. At the present time it is not possible to decide whether such extended features result from the elastic energy available being sufficient to overcome the reduction in stress concentration which occurs when a propagating microcrack enters a pore, thus enabling propagation to continue, or whether they arise from the overlap and joining together of newly generated and previously existing microcracks.

Failure is considered to occur when the concentration of microcracks and extended microcracks becomes so high that the generation formed by the next increment of the load results in the joining together of sufficient stable microcracks to form a large unstable flaw. Then, according to simple flaw theory, failure directly ensues. Graphites, despite having quite different structures, also fail by a mechanism involving the formation of unstable flaws from the stable microcracks induced at lower stress levels [8].

On this failure mechanism a high sub-critical microcrack density is a precondition for failure. Since the as-received specimens exhibit a wide variation in the number of microcracks they contain, it is reasonable to postulate that the dispersion of tensile strength values for a coke is related to the flaw density in the individual as-received specimens. On this basis those as-received specimens in Fig. 8 which contain a high microcrack density would be likely to fail before the mean load was attained. Thus data for the "stressed" specimens are more comparable with those for the as-received specimens containing fewer flaws.

Fracture crack systems in broken specimens are very complex (Fig. 7). The multiplanar and branched cracking observed results from the strain energy released exceeding that necessary merely to propagate the crack and therefore initiating secondary cracks ahead of the propagating tip. However, some contribution to the complexity of the fracture crack system can be expected to arise from secondary cracking near the loading positions. There is some evidence in Fig. 7 of cracks, usually subsidiary ones, occurring at high angles to the loaded diameter, and in such cases it is possible that shear forces were involved in their propagation. Nevertheless, Fig. 7 does show that in general the broken specimens do contain a diametral fracture plane. This is one criterion necessary for the determination of a valid tensile strength value by this technique [9]. However, previous work [10] showed that when industrial coke specimens were loaded by diametral compression marked deviations from the theoretical stress distribution occurred, so that the tensile strength values should be considered as comparative measurements.

The tensile strength of coke has been related statistically to the mean Feret's diameter of the larger pores, the form of the equation implying that the larger pores act as the critical Griffith flaws in coke [2]. Clearly the larger pores control the coke tensile strength. Furthermore they appear to provide the stress concentration necessary for the initiation of microcracks. However, the direct evidence now available demonstrates that there exist in coke, prior to its failure, extended microcracks of linear dimension substantially greater than the maximum dimension of the larger pores. Perhaps therefore the critical Griffith flaws should now be regarded as consisting of the series of pores and interconnecting microcracks which constitute these extended microcracks.

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