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THE EFFECT OF MACERAL COMPOSITION ON THE  
BINDERLESS BRIQUETTING OF HOT CHAR

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## 1. INTRODUCTION

The process of hot char briquetting now being developed and exploited by the National Coal Board consists of the direct briquetting of hot fluid-bed carbonised char. The principal process variables have already been discussed by Habberjam and Gregory (1), and this paper concerns an extension of their work.

The coal seams so far considered for the hot char briquetting of low rank coal have been of similar petrographic composition, i.e. about 60% vitrinite, 17% exinite, and the remainder composed of micrinite 1 and 2, fusinite, shale and pyrite. There is no reason why other coal seams should not be examined, particularly since certain coal preparative treatments, e.g. selective froth flotation and dense medium separations, have the effect of concentrating one particular maceral or group of macerals at the expense of the others. It was considered that coals of unusual maceral composition might exhibit a new range of briquetting properties.

Although work has been published on the thermal changes occurring in individual coal macerals, no systematic survey has been made using British low rank coals heated in fluid-bed conditions.

Many workers, notably Krüger (2) and Fitzgerald (3) have shown the pyrolysis behaviour of the individual coal macerals to be different. They, as well as Permittina (4) and Amosov (5), were working under coke oven conditions; Permittina, using polished microscope sections of Russian coking coal chars, showed that only vitrain, and vitrainised masses and spores, contributed significantly to caking behaviour. Permittina also showed that fusainised micro-components, xylene\* and minerals, made no contribution, whilst xylo-vitrain and partially fusainised attrition particles had low caking properties. Ergun (6), working with a microscope hot-stage, has confirmed that vitrinites and exinites undergo visible changes during carbonisation, and that the other constituents remain comparatively inert. Taylor (7) studied the thermal properties of the Stopes concentrates during carbonisation, and lists a considerable quantity of relevant literature.

In this work the maceral system of coal component classification will be used. The components analysed will be:

vitrinite, exinite, total inertinite, shale and pyrite.

The inertinite will be further divided into micrinite 1, micrinite 2, and fusinite.

\* The word "xylene" is taken directly from a translation of Permittina's paper. It probably implies xylo-fusain.

The detailed objects of the present study were:

- (a) to devise a method for estimating volume changes in the macerals of low rank coal chars heated in a fluid bed;
- (b) to show whether variations in the maceral compositions of the coal feed would affect the char briquetting process;
- (c) to follow any changes occurring in the macerals of chars in the range 380° to 500°C.

## 2. EXPERIMENTAL

### 2.1. Coal sample preparation

Samples of four low rank, high volatile coals were used. An analysis of these coals is given in Table 1.

TABLE 1  
Analyses of coals used

Colliery	Seam	Commercial grade	Moisture % as received	Ash % as received	Volatile matter d.a.f.
Calverton	High Main	Washed smalls	7.59	11.68	36.83
Denby Hall	Mixed	Washed smalls	5.25	16.50	37.80
Birch Coppice	Mixed	Washed smalls	5.53	11.11	41.19
Dexter	Mixed	Washed special beans	6.18	1.66	38.03

Samples of one coal, namely Calverton CRC.902, were used to prepare maceral concentrates. Hards, brights and fusinised materials were manually selected, and further refined by flotation and crushing, followed by selection under a low-power lens. Three basic samples were prepared in this way, the proximate analyses being given in Table 2.

TABLE 2  
Proximate analyses of basic samples

	Durain	Vitrain	Fusain
Moisture	5.99%	12.47%	4.55%
Ash (dry basis)	7.67%	1.07%	10.29%
Volatile matter (d.a.f)	38.49%	35.61%	24.80%

Ten mixes were prepared from the three basic samples, the vitrain, durain and fusain samples being listed as Sample Nos. 1, 8 and 10 respectively (Table 3).

TABLE 3

Analyses of Calverton coal  
maceral concentrate blends

Sample No.		1	2	3	4	5	6	7	8	9	10
Vitrinite	Vol. %	97.9	74.7	51.4	67.4	50.9	33.3	35.7	14.5	16.7	23.9
Exinite	"	1.7	9.8	28.6	5.7	21.0	33.2	14.8	51.1	34.4	5.4
Total inertinite	"	0.4	15.5	19.3	26.9	27.7	33.0	49.3	34.4	48.5	70.3
Micrinite 1	"	0.3	7.5	12.7	14.7	18.6	17.9	27.2	26.8	31.0	33.7
Micrinite 2	"	0.1	4.8	6.5	7.8	7.4	12.2	15.2	7.3	12.1	24.3
Fusinite	"		3.2	0.1	4.4	1.7	2.9	6.9	0.3	5.4	12.3
Shale	"			0.7		0.3	0.5	0.2		0.4	0.1
Pyrite	"					0.1					0.3

## 2.2. Apparatus

Two systems of fluid-bed carbonisation were used:

- (1) A 2-inch laboratory-scale fluid-bed of the design described by Habberjam and Gregory (1). This apparatus was used for the study of the pyrolysis of macerals. Two carbonisation periods were used in this fluid bed, i.e. 8 and 30 min., the first 4 min. of each period being the heating time. Nitrogen was used as the fluidising gas.
- (2) A miniature fluid-bed and briquetting mould. This apparatus consisted of a 0.5-inch diameter tapered mould and hardened steel plunger mounted in the jaws of a hydraulic jack. A tightly fitting funnel was placed in the top of the mould, so that the mould and funnel together acted as a fluid-bed, the funnel being removed from the mould for briquette production. Air and nitrogen were used as the fluidising gases.

One carbonisation period was used in the miniature bed, i.e. 5 min., including a 2 min. heating period.

## 2.3. Testing and analysis of samples

The briquettes prepared at 6 tons/sq.in. were tested for their bulk density and mechanical strength by the method described by Habberjam and Gregory (1).

The samples for maceral analysis were ground as coal to -10 B.S.S. and reduced to 30 gm. using an Otto Microsplitter (8). These samples were then carbonised in the 2-inch fluid-bed, the chars being further reduced to approximately 1 gm. samples before impregnation and mounting in blocks with cold-setting polyester resin. A flat surface was ground on the blocks, using several grades of carborundum powder on glass plates, and a high polish was obtained with fine alumina powder on a Bolting silk lap.

A statistical method of analysis, based on a technique first described by Delesse (9) was used to determine the relative volume of macerals in the coals and chars. This was based on the fact that, in a cross-section of rock, the ratio of the area occupied by one mineral to the total cross-sectional area is a reliable estimate of the total volume percentage of that mineral.

Several methods of measuring the relative areas of minerals in cross-section have been proposed, but the most reliable was taken as the 'point counter' technique described by Glagolev (10). In theory, the cross-section is covered with a grid and the mineral occurring under each intersection is recorded. The ratio of the number of points at which the particular mineral occurs to the total number of points of the grid may be reliably taken as the ratio of the area of the mineral to the total measured area. A mathematical proof of these statements is given by Chayes (11). In practice, instead of moving from point to point on a grid, the polished coal section is moved in successive steps along a number of traverses, and the mineral under the crosswires is recorded at each step. In order to limit the error to  $\pm 3\%$  at 75% and to  $\pm 1\%$  at 5%, a total of 1,000 points is analysed on each sample.

In order to estimate the changes in the volumes of the macerals with respect to the original coal, it is necessary to have some body or bodies present which remain inert during carbonisation. Since their volume does not change, comparison is possible with the changing maceral volumes. Where a natural inert material is present in reasonable percentages, this may be used. More than 10% must be present, however, to ensure that the errors are less than the probable error in the maceral analysis.

For the coals Calverton, Denby Hall and Birch Coppice, micrinite 2, fusinite and minerals were regarded as standard inerts. In the case of Dexter, a percentage of gas coke ground to -10 B.S.S. was added to the original coal charge. The porosities, also found by a 'point counter' method, are a derived figure (see section 3, Discussion). Sections of briquettes were also mounted in resin, and photomicrographs of sample chars were made with a Vickers projection microscope. (See photos. 1 through 12.)

### 3. RESULTS AND DISCUSSION

The experimental conditions for briquetting and for maceral analysis were unavoidably different. The briquetting trials using the very small quantities of sample available were carried out using a miniature fluid bed which allowed for heating rates of approximately 200°C/min. Three minutes after the attainment of carbonisation temperature, i.e. 5 min. after sample introduction, the briquettes were formed. In the maceral analysis larger samples were available, and a more convenient and conventional fluid bed was employed. The rate of heating in this bed was approximately 100°C/min. and samples were analysed 4 min. and 26 min. after they had attained carbonisation temperature, i.e. total residence times of 8 and 30 min. respectively. Previous work had shown that briquettes formed in the miniature fluid bed after heating for 5 min. had a similar performance to those after 8 - 10 min. in the more conventional bed.

The results of the briquetting of maceral concentrates are given in Figures 1 to 3. Calverton coal (C.R.C. 902, V.M. 37% d.a.f.) was used and the three methods of briquette assessment employed, i.e. shatter index, abrasion index and bulk density, gave similar results. Maceral concentrates rich in inertinite produced poor briquettes, whilst both exinite and vitrinite rich mixtures produced good briquettes. Exinite rich mixtures containing over 25% micrinite 1 produced good briquettes, whereas inertinite rich mixtures with the same concentration of micrinite 1 produced poor briquettes. It was therefore unlikely that micrinite 1 contributed to the production of strong briquettes. An interaction between exinite and micrinite was possible and this was not ruled out by the microscopic study of the macerals (Fig. 4). Micrinite 1 underwent some swelling between 375° and 400° C, the swelling being greater after 30 min. carbonisation than after 8 min. This swelling occurred for all

the coals considered (Figs. 5 and 7).

Samples containing practically pure vitrinite (97.9% by volume) produced strong compacts throughout the range of temperatures at the short carbonisation time studied, the shatter and abrasion strengths falling slightly at the higher temperatures. A photomicrograph of a 97.0% "pure" vitrinite briquette (Photo. 11) illustrates the way in which the vitrinite grains appear to 'jig-saw' into one another. The maceral analyses show a series of expansions and contractions throughout the range which are only qualitatively followed by the other coals studied.

The exinite rich mixtures (up to 51% volume) did not produce strong briquettes before 410°C, but above this temperature the exinite played an important role in briquetting, either alone or in combination with vitrinite or micrinite 1. The quality of exinite rich compacts rose to a maximum at about 440°C and then fell gradually. A photomicrograph (Photo. 12) of a 'whole' coal briquette (exinite 17%, vitrinite 66%, micrinite 17%, and micrinite 2 plus fusinite 10%) shows definite bridging of coal grains by melted exinite. As a maceral the exinite appeared to be lost, both with respect to increasing temperature and carbonisation time. Its disappearance as a recognisable maceral would not preclude its impregnation into the microstructure of remaining coal substance. The changes in the maceral structure were coincident with porosity changes. There was no significant difference for nitrogen and air fluidisation.

The porosities of the chars (Fig. 8) rose steeply with temperature and increased with period of carbonisation. In determining the porosities of chars, three separate types of pores were measured: rounded holes, angular holes and natural holes. It was probable that the rounded holes were formed as the gases resulting from the decomposition of exinite and vitrinite attempted to escape through the plastic or semi-plastic vitrinite, and that the angular holes were cracks formed after carbonisation when the material contracted on cooling. The third classification of holes, natural holes, occurred in fusinite and semi-fusinite grains. The porosities shown in Fig. 8 are 'rounded hole' porosities.

Table 4 below illustrates the relative importance of the rounded, angular and natural holes in Calverton coal.

TABLE 4  
Porosity Analyses of Calverton Chars

<u>Calverton</u> <u>8 min. N<sub>2</sub></u>	<u>Rounded</u> <u>holes</u>	<u>Angular</u> <u>holes</u>	<u>Natural</u> <u>holes</u>	<u>Total</u> <u>porosity</u>
350°C	0.1	1.6	2.4	4.1
375°C	1.4	1.6	1.8	4.8
400°C	0.7	2.2	2.3	5.2
425°C	21.6	3.9	3.0	28.5
450°C	43.4	3.2	1.1	47.7
475°C	51.7	3.5	1.1	56.3
500°C	60.5	2.6	1.0	64.1
<u>30 min. N<sub>2</sub></u>				
350°C	0.4	2.0	1.1	3.5
375°C	0.4	1.9	1.3	3.6
400°C	10.2	3.8	2.0	16.0
425°C	32.3	4.0	1.9	38.1
450°C	47.3	3.5	1.2	52.0
475°C	52.8	3.4	0.3	56.5
500°C	59.4	2.6	1.2	63.2

Photomicrographs of chars heated in air and in nitrogen illustrate the formations of these holes and the relative changes in the macerals. (See photos. 1 through 10)

On a commercial scale, the results of these investigations illustrated the danger of highly selective coal preparative treatments. Changes of the maceral composition of the coal feed to a char briquetting plant might well affect the operation of the process, e.g. the addition of froth flotation fines would lower overall quality and a change of exinite/vitrinite ratio might well affect the optimum temperature of operation.

On the laboratory scale, the work has illustrated that changes in the volume of individual macerals can be estimated by comparison with naturally inert material in coal, and that results using this method are similar to those using an artificial inert additive.

#### 4. CONCLUSIONS

- (i) The maceral volume changes in coal chars can be estimated by the use of naturally occurring inert material where over 10% is present in the original coal. Where this naturally inert material is not present, an artificial additive, e.g. high temperature coke, may be used.
- (ii) On a commercial scale, sudden or gradual changes of the maceral composition of coal feed to a char briquetting plant may well affect the operation of the process. High inertinite content raw material will lower overall strength, and any change in exinite/vitrinite ratio may affect the optimum operational temperature.
- (iii) For a non-coking high volatile coal, exinite, vitrinite and micrinite 1 were shown to undergo changes in the range 350°C to 500°C. The vitrinite, and to a lesser extent, micrinite 1, swell and contract in this range, whilst the exinite is lost completely as a recognisable maceral.

#### 5. REFERENCES

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CAPTIONS TO PHOTOMICROGRAPHS OF CHAR GRAINSPhotomicrographs of 30 minute Calverton Chars C.P.S.1855/16(Magnification approximately X 120)Photo.  
No.1. 350°C N<sub>2</sub> Fluidisation

The exinite appears unaltered at this temperature. The vitrinite shows cracking around the inert fusinite mass, which indicates that there may be contraction.

2. 350°C Air Fluidisation

The exinite appears unaltered except where it lies on the margins of grains and has been oxidised. The vitrinite is cracked, and has fairly wide oxidised margins extending up the open cracks.

3. 400°C N<sub>2</sub>

The exinite is disappearing, leaving irregular holes in the micrinite and vitrinite. The megaspore in the larger durain grain has become liquid and some of it has migrated to the edge of the grain, cementing a triangular vitrinite grain to it. The micrinite and fusinite are unaltered, and the vitrinite remains angular, with fine cracks.

4. 400°C Air

A large megaspore has become oxidised and has cracked away from the durain grain which contains it. Within this megaspore there is a small area of unoxidised volatile matter which is boiling off. There is unoxidised exinite boiling off in both durain grains. In the triangular grain the megaspores have been partially oxidised and the transition between volatile and non-volatile exinite is shown. The vitrinite is just beginning to become plastic enough for rounded bubbles to develop in some of the grains.

5. 425°C N<sub>2</sub>

Most of the exinite has disappeared. The vitrinite grains are becoming plastic, and some grains are filled with gas bubbles. The inertinites are still unaffected, except by escaping from enclosed vitrinite and exinite.

6. 425°C Air

The vitrinite appears to be considerably more plastic than in N<sub>2</sub> at this temperature. The grains are considerably swollen and their margins are rounded. There is still some exinite present in the durain grain. The inertinites remain as in the 400°C sample. It can be seen that the margins of the vitrinite grains are still sealed, since the bubbles have no oxidation rims, indicating that the margins must still be quite plastic.

7. 450°C N<sub>2</sub>

The large durain grain has remained inert, except for a vitrinite band which is boiling out. The microspores and megaspores have gone, leaving a skeleton of micrinite, which has not even collapsed into the space left by a megaspore. The pure vitrinite grains are swollen, but the ones shown in this micrograph still have fairly thick walls.

8. 450°C Air

Again the decomposition of vitrinite appears to be in a later stage than in the 450°C N<sub>2</sub> chars. Some of the bubbles contain oxidised margins, and some are unoxidised, being still enclosed.

9. 500°C N<sub>2</sub>

The vitrinite grains have become quite rounded, consisting of a network of thin-walled bubbles. Bands of micrinite 2 have remained with their original structure.

10. 500°C Air

The stage reached is very similar to that of the N<sub>2</sub> char at this temperature, except for the oxidation of the grain and pore margins. Again, the micrinite 2 and fusinite appear to have its original structure. Most of the bubbles have been oxidised, showing that the surface of the grains has been broken, and that the oxidised vitrinite has become brittle at this temperature.

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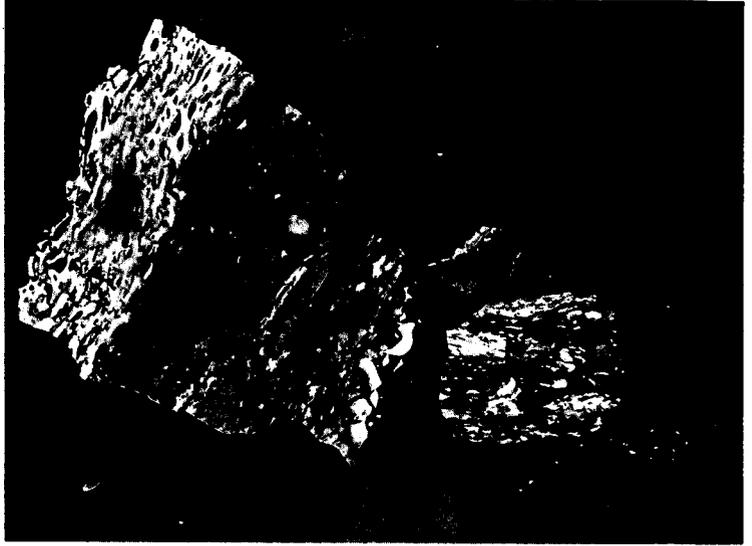
1. 350°C N<sub>2</sub>



2. 350°C Air



3. 400°C N<sub>2</sub>



4. 400°C Air



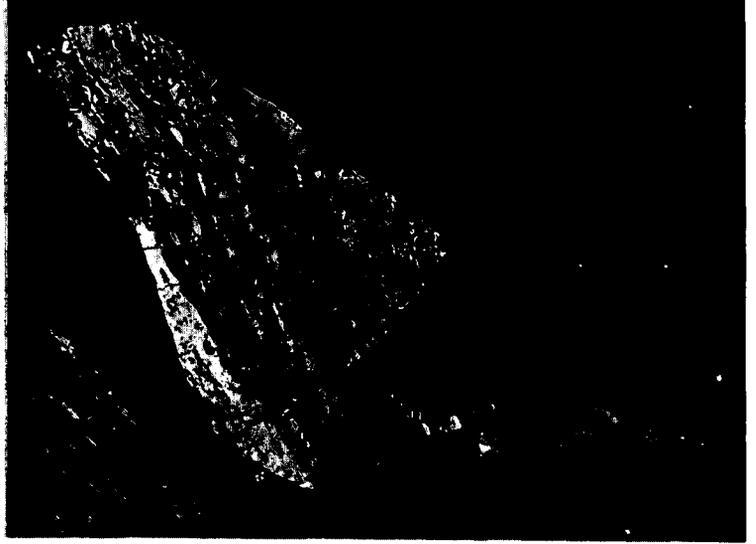
5. 425°C N<sub>2</sub>



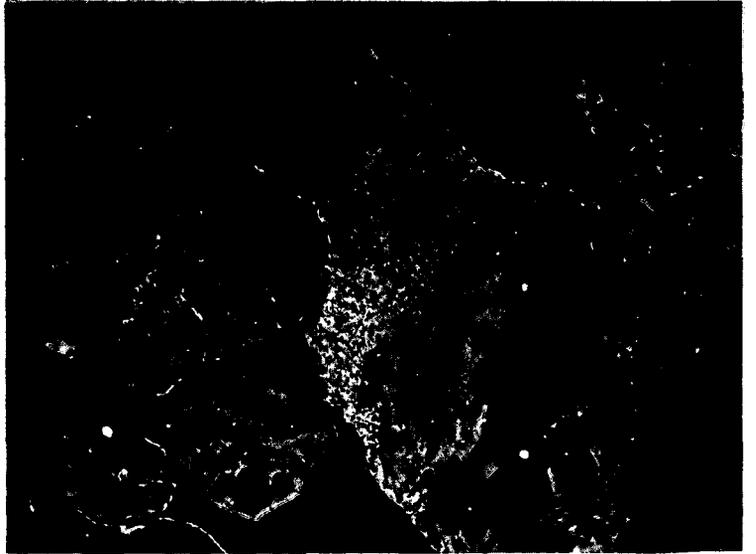
6. 425°C Air



7. 450°C N<sub>2</sub>



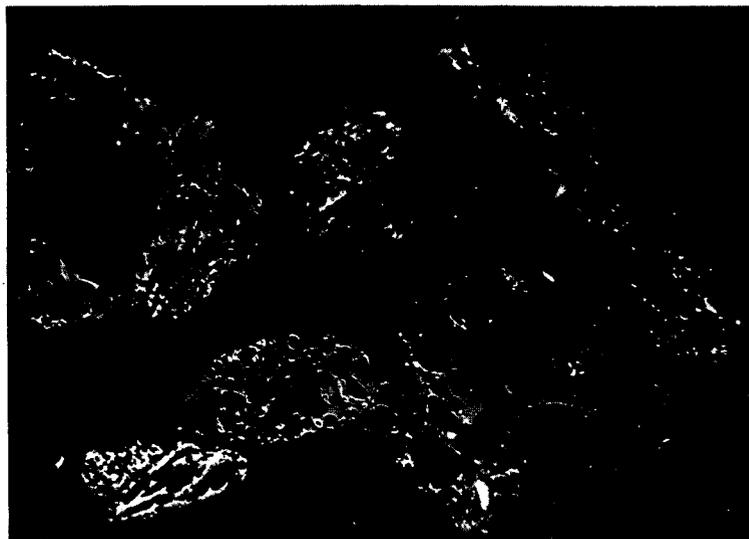
8. 450°C Air



9. 500°C N<sub>2</sub>



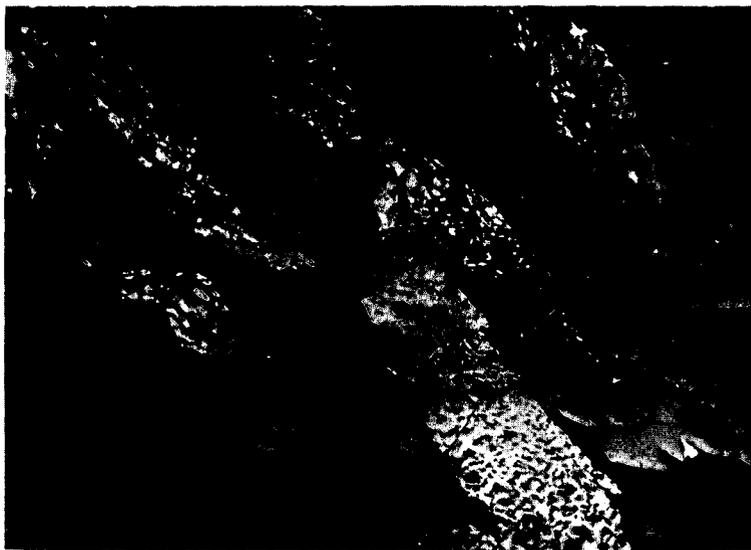
10. 500°C Air



11. A 420°C  
"pure"  
vitrinite  
briquette



12. A 420°C  
"whole coal"  
briquette



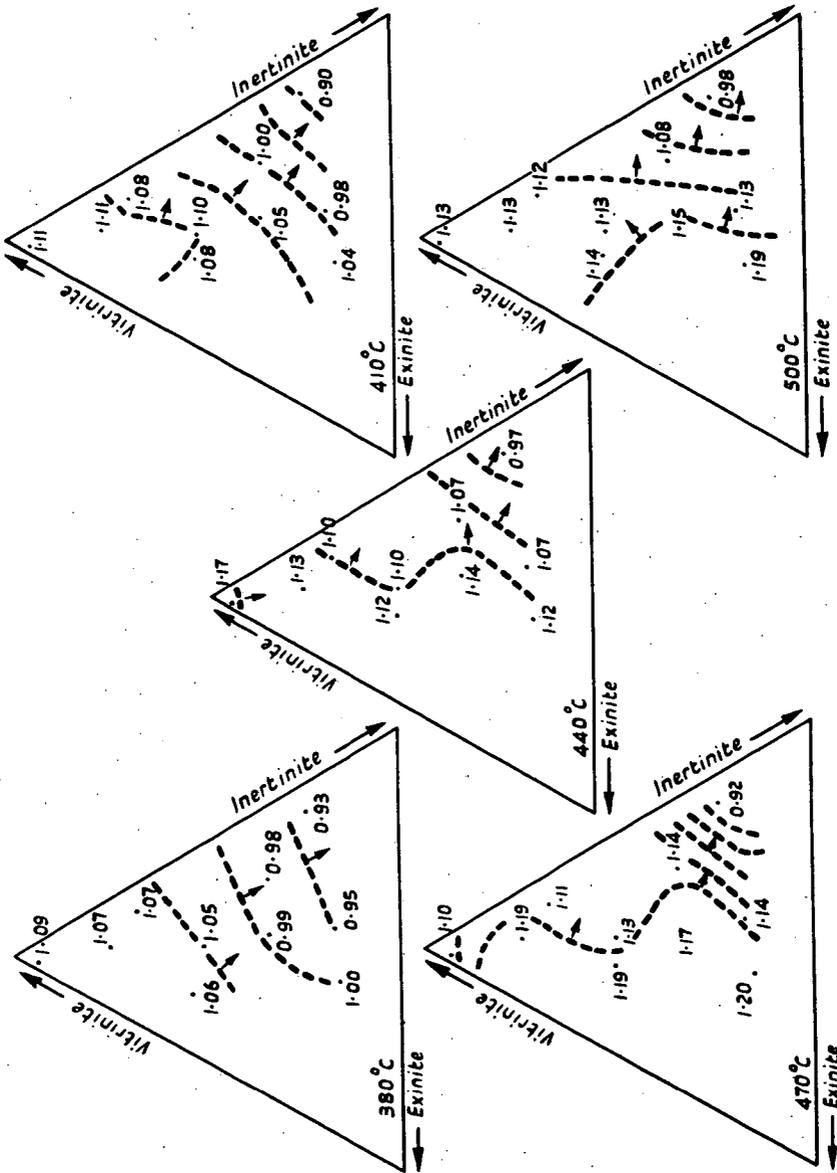


FIG. I. DENSITY OF CHAR BRIQUETTES, g/cc.

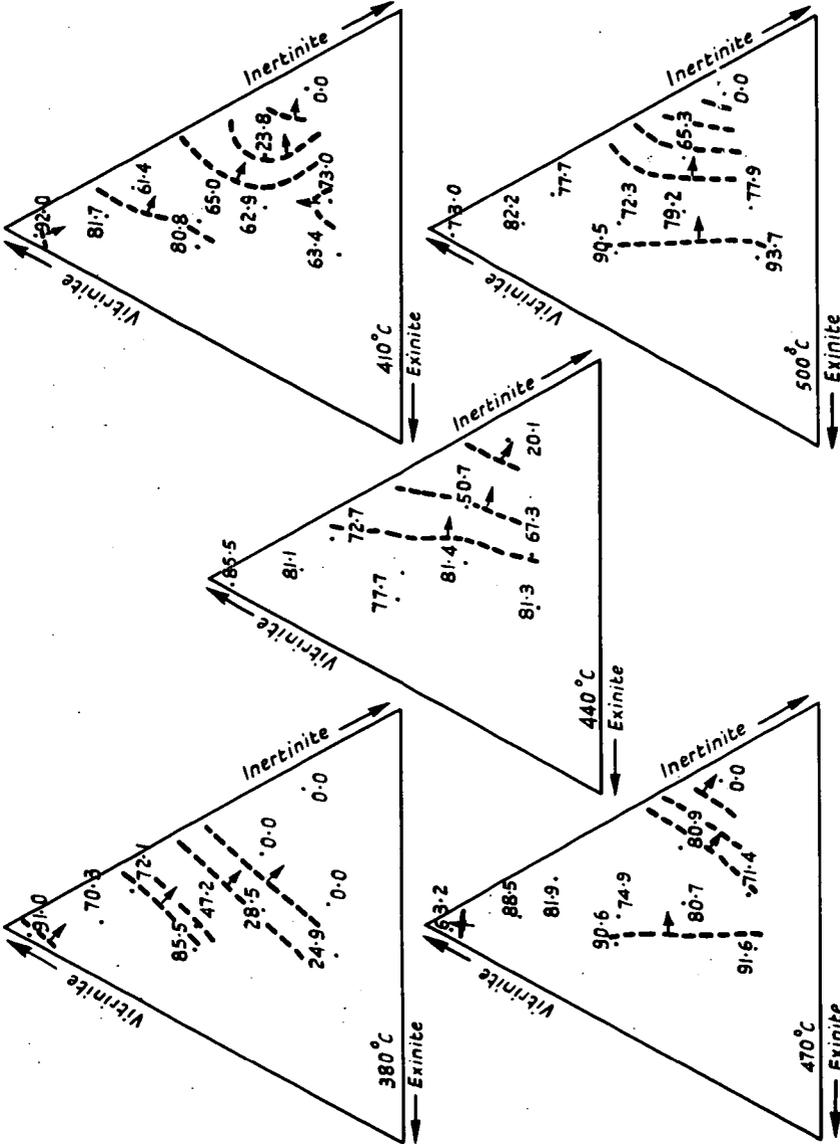


FIG. II. VARIATION OF ABRASION INDEX WITH TEMPERATURE AND COMPOSITION.

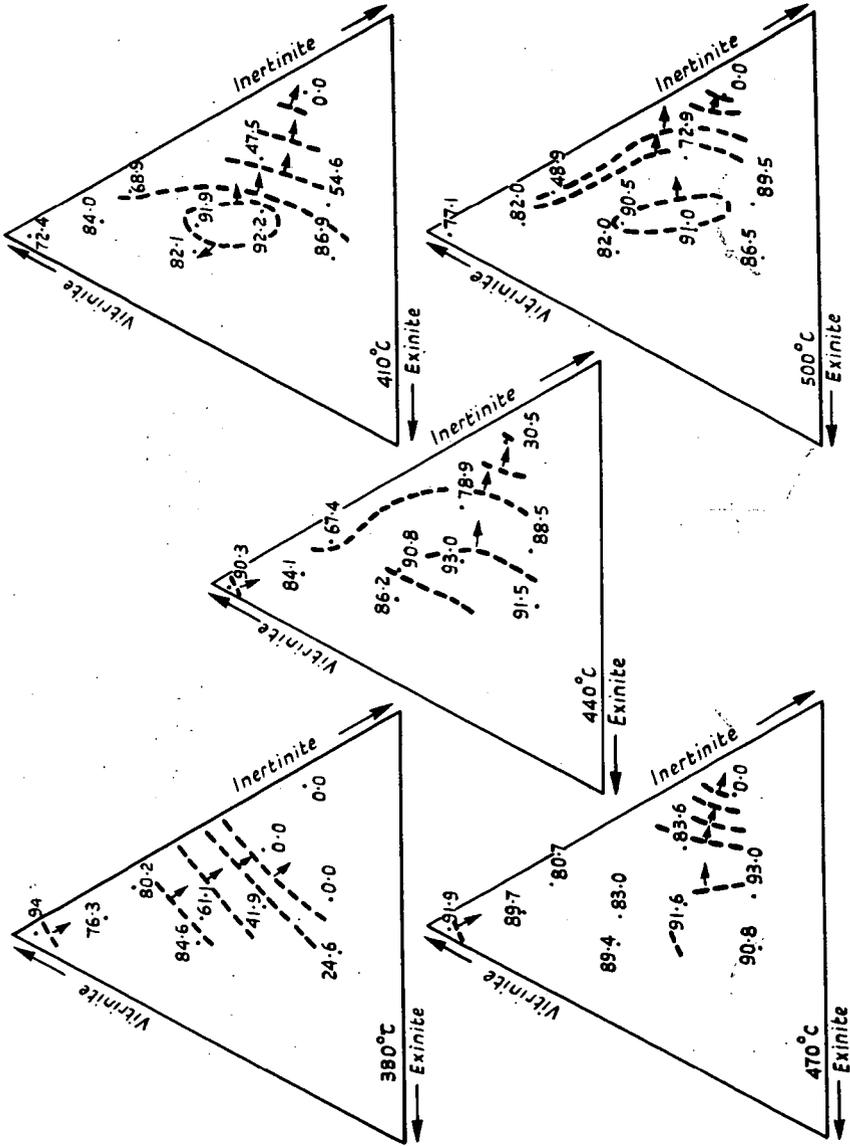


FIG. III. VARIATION OF SHATTER INDEX WITH TEMPERATURE AND COMPOSITION

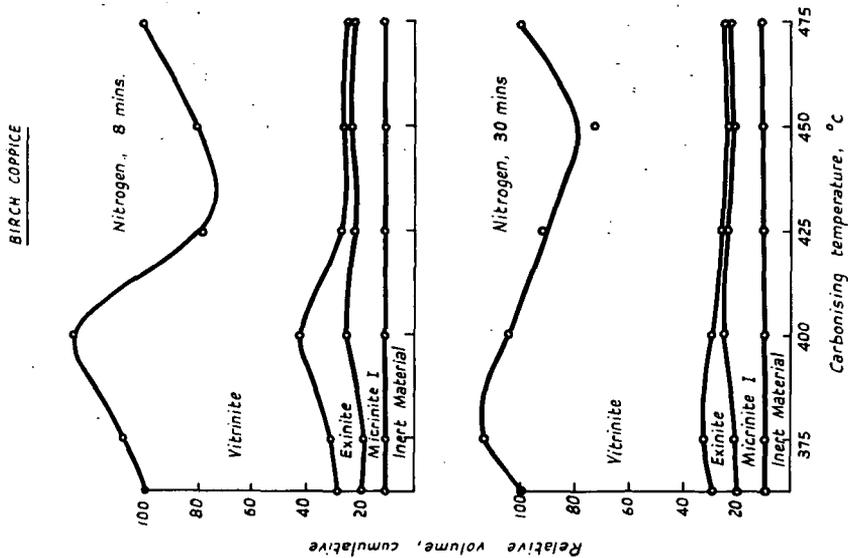


FIG. I RELATIVE VOLUME OF MACERALS.

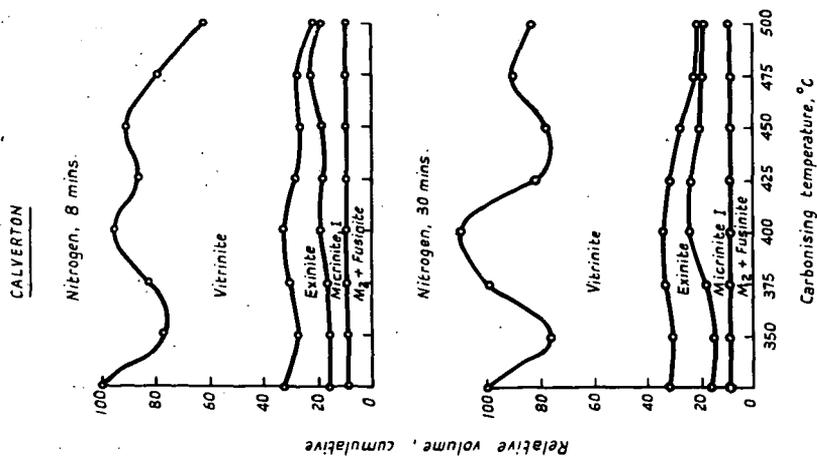


FIG. IV. RELATIVE VOLUME OF MACERALS.

DENBY HALL

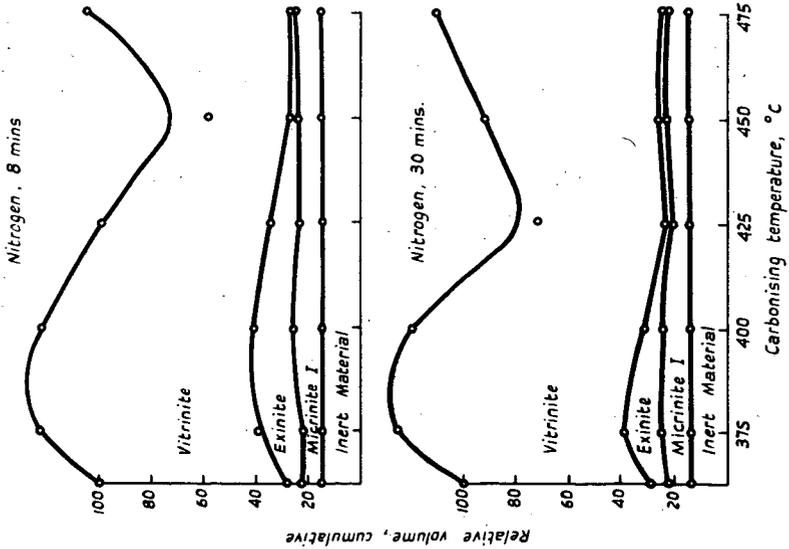


FIG. VI. RELATIVE VOLUME OF MACERALS.

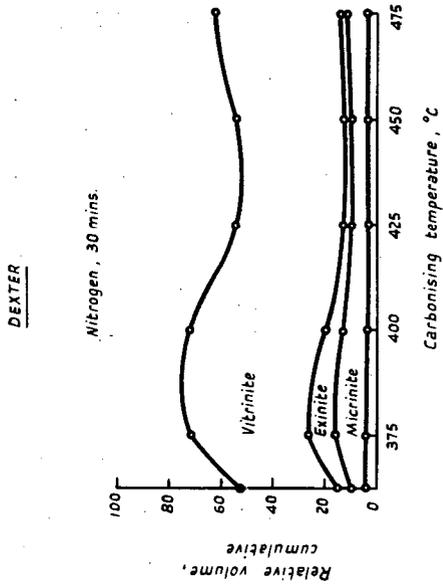


FIG. VII. RELATIVE VOLUME OF MACERALS.

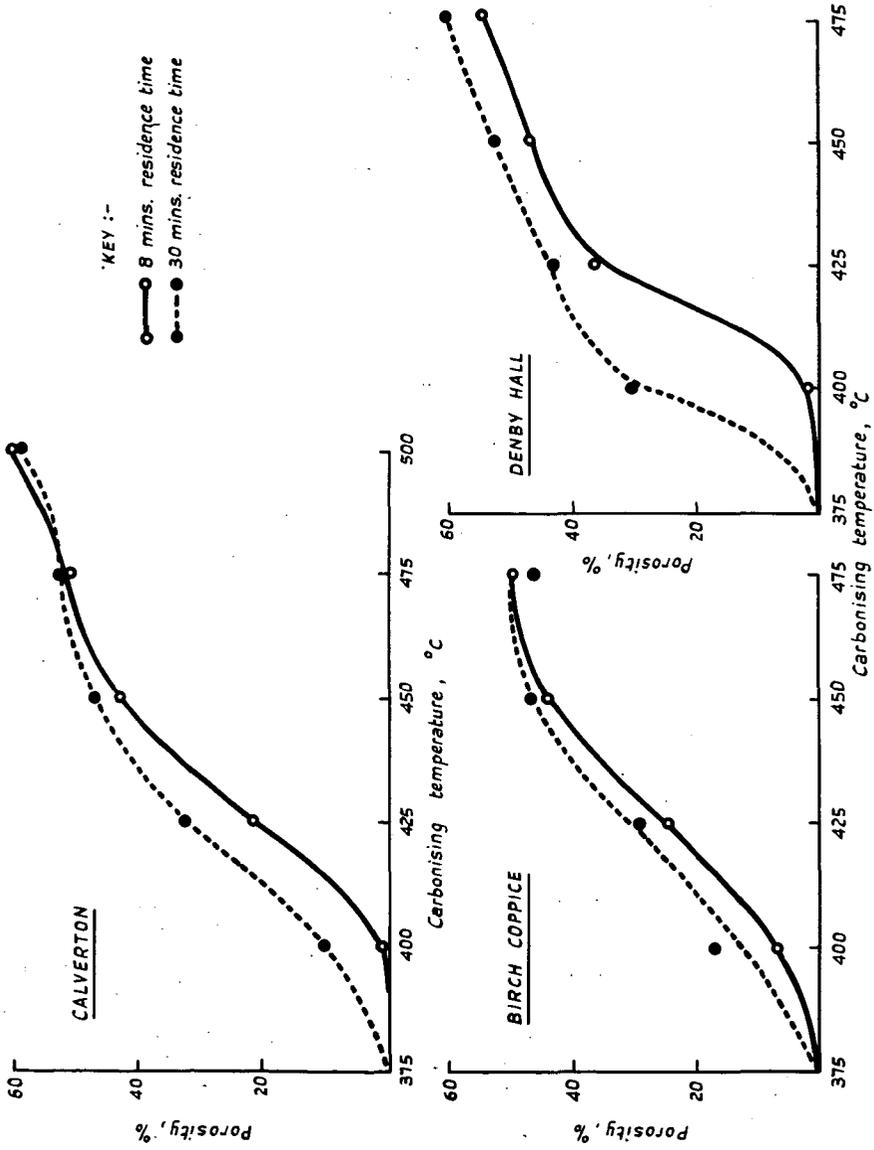


FIG. VIII. POROSITY OF CHAR FROM THREE LOW RANK COALS.