

BEHAVIOR OF CLAYS ASSOCIATED WITH LOW-RANK COALS
IN COAL-CLEANING PROCESSES

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About 65 percent of the 400 plus million tons of coal produced in the United States annually is mechanically cleaned to remove the inorganic impurities before being sent to market. The removed impurities, principally shale and clay with some pyrite, average about 20 percent of the raw coal treated. Nearly 95 percent of the tonnage treated is dealt with by wet processes in which water, suspensions of solids in water, or occasionally calcium chloride solutions are employed. Difference in density between coal and impurity is the basis of these processes.

Of major importance in the separation is the competency of the impurities, that is, the resistance of the impurities to size disintegration in water. In general, the impurities are more resistant in geologically older coals or those in effect made older by metamorphism. If the impurities suffer reduction in particle size when immersed in water, the separation of organic from inorganic material becomes more difficult. Recently the Scientific Department of the National Coal Board (Great Britain) undertook a fundamental investigation of the interaction of shale and water and also developed an empirical method of assessing shale breakdown or disintegration (1).

Comparatively little is known about the behavior of the clay and shale associated with subbituminous coal and lignite because experience in upgrading them by the more efficient wet-beneficiation methods is limited. These lower-rank coals occur in the western states where energy requirements in the past have been relatively small, and supplies of alternate fuels have been abundant. Now however, these coals, which comprise the bulk of the Country's reserves of mineral fuel, are becoming of interest as sources of fuel for power generation and as potential sources of metallurgical carbon and carbonization byproducts. Thus, their behavior in preparation processes may soon become important.

The more common tendency of geologically younger shale associated with low-rank coal to disintegrate and form slimes or suspensions in the wet-cleaning operation has been examined in our coal-preparation laboratory during the treatment of a subbituminous coal and a lignite from Washington and a subbituminous coal from Oregon. Not only was it found that particle-size disintegration through softening and dispersion of colloidal matter was of importance, but also that the lattice structure of the particular clay minerals which rendered it susceptible to swelling and consequent reduction in apparent density was of great significance.

Thermal Analysis

Differential thermal analyses (DTA) of the principal interbedded impurities in the three coals are shown in figure 1. These data characterize all of the clays as bentonitic, with sodium and calcium montmorillonite minerals predominating. One characteristic property of montmorillonites is their extremely small particle size and hence the ability to form fairly stable suspensions in water.

Interpretation of the DTA curves in the figure is considered to characterize sample 1, from the Big Dirty Bed, Lewis County, Wash., as principally sodium montmorillonite; sample 2 from the Southport bed, Coos County, Oreg., contains mainly calcium montmorillonite with some illite; and sample 3, the lignite from near

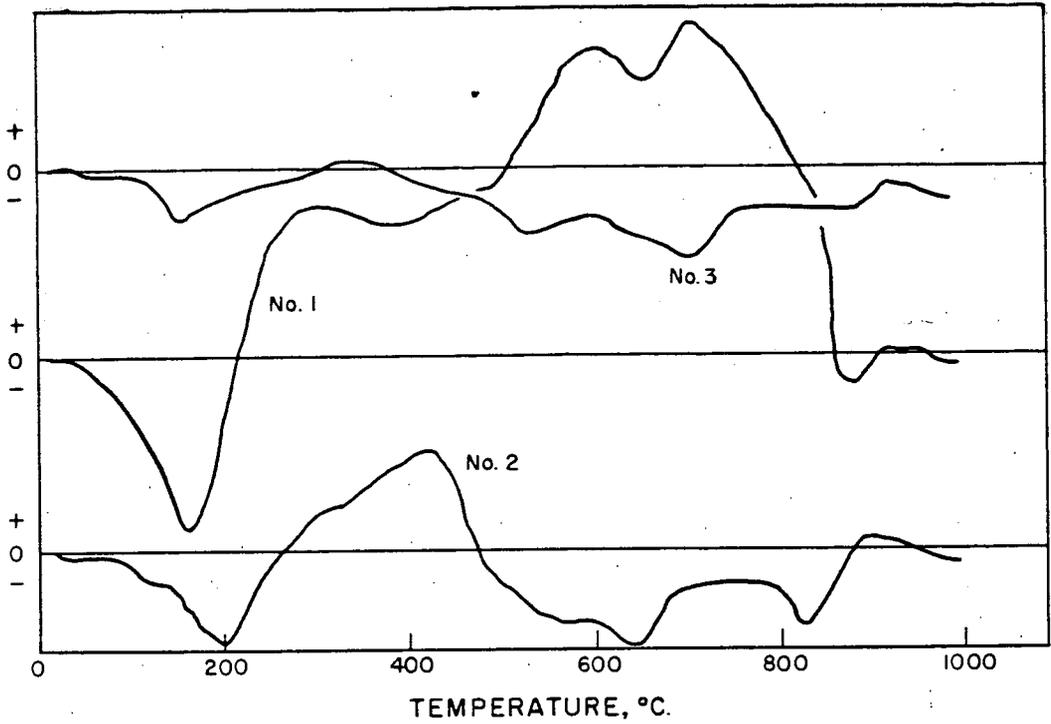


Figure 1. Differential-thermal-analysis curves.

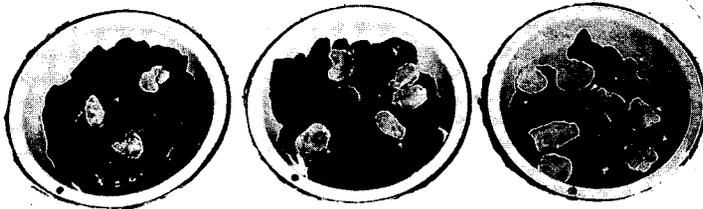


Figure 2. (Left to right). Swollen clay in 1.50 to 1.60, 1.60 to 1.70, and 1.70 to 1.80 density fractions of subbituminous coal from Lewis County, Wash., size 3/4 to 1 1/2 inches.

Swelling is due to water penetration between the lattice layers. The following tabulation shows the extent of relative swelling of the principal impurities of the three coals, as determined by a commonly used swelling test (2).

<u>Sample No. and source</u>	<u>Volume increase, percent</u>	
	<u>6 hours</u>	<u>24 hours</u>
1. - Big Dirty bed, Wash.	100	114
2. - Southport bed, Oreg.	75	75
3. - Toledo lignite, Wash.	25	25

The clay from the Big Dirty bed was hand picked from a 1.5 to 1.6 specific gravity fraction. Its swelling illustrated the magnitude of the density change that might be encountered in cleaning young coals, and also explains why clay was present in the 1.5 to 1.6 fraction.

Washington Subbituminous Coal

The first coal examined was from the Big Dirty bed in Lewis County, Wash., a deposit with sufficient reserves of relatively low-cost coal to be of interest for onsite power generation, despite its low grade. It is subbituminous B in rank, and in the raw condition has the following analysis:

	<u>As received</u>	<u>Moisture-free</u>
Moisture, percent	20.0	----
Volatile matter, percent	28.6	35.8
Fixed carbon, percent	27.9	34.8
Ash, percent	23.5	29.4
Sulfur, percent	0.6	0.8
B.t.u., per pound	7240	9050
Ash-softening temperature, °F.	----	2250

The fresh coal is strong, resists degradation, and has a Hardgrove grindability index of 35 to 40, depending on its degree of freedom from impurity. If allowed to dry, however, the coal rapidly develops shrinkage cracks and becomes quite friable.

Washability Examination. A 28-ton sample of coal was obtained for examination, about 3 tons for specific-gravity analyses, and the rest for cleaning trials. Special precautions were observed in preparing the samples for float-and-sink tests to insure against drying, which would have altered the particle size of both the coal and the clay. All sizes of coal coarser than 20 mesh were tested in aqueous solutions of zinc chloride, using each 0.1 interval in specific gravity from 1.30 to 1.80. Material finer than 20 mesh was air-dried and was tested in organic liquids of the same densities.

Of special interest in these analyses, shown in table I, was the abnormally high ash contents of the specific-gravity fractions between 1.40 and 1.80, particularly in the coarser sizes, occasioned by the swelling nature of the clay. For example, in the 4- to 2-inch size, the 1.70 to 1.80 fraction analyzed 75.4 percent ash, whereas about 50 percent ash would be normal; similarly, in the next lighter fraction, the ash content was 65.9 percent, instead of the usual 40 to 45 percent. These high ash contents confirmed the visually observed anomaly that free pieces of clay, derived from partings within the coal bed, were present in all specific-gravity fractions heavier than 1.40. Figure 2 is a photograph of several of the density fractions of the 3/4- to 1 1/2-inch size, showing the presence of clay.

TABLE I. - Specific-gravity analyses of raw coal from Big Dirty bed

<u>Size, inches</u>	<u>Specific gravity</u>	<u>Weight-percent</u>	<u>Ash, ^{1/}percent</u>	<u>Cumulative ^{1/}</u>	
				<u>Weight-percent</u>	<u>Ash, percent</u>
4 to 2 Weight, 22.2 percent	Under 1.30	10.4	6.7	10.4	6.7
	1.30 to 1.40	59.5	13.5	69.9	12.5
	1.40 to 1.50	12.1	32.5	82.0	15.4
	1.50 to 1.60	6.0	54.7	88.0	18.1
	1.60 to 1.70	3.8	65.9	91.8	20.1
	1.70 to 1.80	1.8	75.4	93.6	21.2
	Over 1.80	6.4	80.9	100.0	25.0
2 to 1/4 Weight, 58.2 percent	Under 1.30	8.3	6.4	8.3	6.4
	1.30 to 1.40	56.1	11.8	64.4	11.1
	1.40 to 1.50	13.3	28.7	77.7	14.1
	1.50 to 1.60	5.4	43.9	83.1	16.1
	1.60 to 1.70	2.6	57.8	85.7	17.3
	1.70 to 1.80	2.0	70.6	87.7	18.5
	Over 1.80	12.3	84.3	100.0	26.6
1/4 to 0 Weight, 19.6 percent	Under 1.30	1.7	6.3	1.7	6.3
	1.30 to 1.40	45.2	8.4	46.9	8.3
	1.40 to 1.50	12.6	19.6	59.5	10.7
	1.50 to 1.60	6.9	29.5	66.4	12.7
	1.60 to 1.70	3.9	39.3	70.3	14.1
	1.70 to 1.80	2.4	47.7	72.7	15.2
	Over 1.80	27.3	81.9	100.0	33.4

1/ Moisture-free basis

from the 4- to 2-inch size fraction and were tested individually in solutions of zinc chloride, with the results shown in the following tabulation.

<u>Specific gravity</u>	<u>Weight-percent</u>	<u>Moisture-free ash, percent</u>
1.40 to 1.50	18.8	52.9
1.50 to 1.60	22.9	72.6
1.60 to 1.70	16.3	85.8
1.70 to 1.80	10.9	88.3
Over 1.80	31.1	89.1

The ash content of 52.9 percent for the lightest particles showed that imbedded coal was present, but the high ash contents of the other density fractions indicated that they were essentially pure clay.

Cleaning Trials With Jig. The jig employed in the cleaning trials was a scale model of a full-size Baum-type jig in every respect except that it had separate elevators for the draw and hutch refuse. The draw product was removed from the upper surface of the screen, and the hutch passed through perforations in the screen. The jig was a 3-cell, single-compartment unit having a nominal capacity of 5 to 6 tons per hour.

The jig was adjusted to remove only the impurity of over 1.65 specific gravity. Operation was entirely on fresh water, all of which was metered. Except for the negligible proportion entrained in the refuse products, all this water left the

system as the overflow of a 6- by 20-foot drag-settling tank in which the washed coal was dewatered; this overflow was sampled continuously to determine the amount and nature of the suspended solids.

The amount of coarse clay observed entering the jig in the feed compared with that leaving in the draw refuse made it evident that much of the clay was being disintegrated in the bed and either was reporting to the hutch or leaving as finely divided solids suspended in the water. Occasional particles of clay, ranging up to 1/2-inch size, were observed in the washed coal.

The following tabulation shows the material balance for the trial and the quality of the products:

<u>Product</u>	<u>Weight-percent</u>	<u>Moisture-free ash, percent</u>
Washed coal	76.4	16.9
Draw refuse	7.5	72.4
Hutch refuse	5.0	68.4
Slimes	11.1	73.7
Total	100.0	29.9

The slimes, that is, the solids suspended in the water, amounted to 11.1 percent of the jig feed, nearly equaling the combined amount of the draw and hutch refuse. The high ash content of these solids indicated they were predominantly clay.

The effectiveness of the jig in removing impurity is shown in the following tabulation:

<u>Size, inches and mesh</u>	<u>Sink 1.70 in washed coal, percent</u>
2 to 1/4	0.5
1/4 to 20	2.4
20 to 0	31.1

Although the removal of impurity from the size coarser than 1/4 inch was excellent, and elimination in the 1/4-inch to 20-mesh fraction was fair, the washed coal finer than 20 mesh was badly contaminated with clay. Many clay particles that did not disintegrate fine enough to become suspended in the water, and were not coarse enough to be drawn down into the refuse bed, adhered to coal particles or were mechanically entrapped in the coal.

The recovery efficiency of the cleaning trial was 96 percent; that is, the yield of washed coal amounted to 96 percent of the theoretical yield of coal of that ash content shown to be present in the feed by specific-gravity analysis. This efficiency is rather low for a separation at 1.65 specific gravity, and is attributable largely to the poor elimination of clay in the finest sizes.

Cleaning trial With Dense-Medium Pilot Plant. The dense-medium pilot plant, in which a suspension of magnetite in water is used, comprises a 24- by 30-inch drum-type separating vessel, a 12-inch densifier, a 12-inch magnetic separator, and a 26-inch by 9-foot vibrating screen, together with necessary pumps and conveyors for handling materials, all arranged in a conventional flowsheet. The separating vessel is a scale model of a widely-used commercial unit. With a 4-ton-per-hour feed rate and the medium flow employed in the tests, the concentration of coal in the separating bath and the retention time were roughly comparable to those in full-scale equipment.

The feed for this cleaning trial was 2- to 1/4-inch in size. Difficulty was experienced in screening at 1/4 inch, even though strong water sprays were used on the vibrating screen. The sticky nature of the clay precluded effective removal of undersize. The material balance for the trial follows:

<u>Product</u>	<u>Weight-percent</u>	<u>Moisture-free ash, percent</u>
Washed coal	82.9	18.1
Refuse	9.0	79.0
Magnetic-separator tails	8.1	71.8
Composite feed	100.0	27.9

The amount of magnetic-separator tailing was considerably higher than generally encountered because of the poor feed preparation mentioned earlier and the unstable nature of the clay.

At the start of the trial the medium, which had a specific gravity of 1.69, contained 7 percent of nonmagnetic material and had a viscosity of 5.3 centipoises. At the end it contained 25 percent of nonmagnetic material and its viscosity was 7.7 centipoises. In a commercial plant unusually high medium-cleaning capacity would have to be provided to maintain medium of suitable quality.

Numerous pieces of clay were observed in the washed coal. Although most of them were finer than 1/2 inch, and many were smaller than the 1/4-inch bottom size of the feed, some plastic pieces coarser than 1 inch were present. The clay was present in larger amounts and coarser size than in the washed product from the jig. The layer of float material in the dense-medium bath is relatively quiescent and therefore soft pieces of clay that would be disintegrated by the shearing and scrubbing action in a jig bed passed over the dense-medium bath in their original form. A specific-gravity analysis of the clay contaminating the washed coal (removed by hand picking) is shown in the following tabulation.

<u>Specific gravity</u>	<u>Weight-percent</u>
1.30 to 1.40	3.4
1.40 to 1.50	6.7
1.50 to 1.60	13.3
1.60 to 1.70	35.0
1.70 to 1.80	25.9
Over 1.80	15.7

The recovery efficiency for this test was 99.0 percent, indicating that there was little loss of coal in the refuse product. In comparing this efficiency with that of the jig, however, it must be borne in mind that the jig was treating material down to 0 size, whereas the dense-medium equipment only treated material coarser than 1/4 inch.

Washington Lignite

Another deposit investigated because of its potential for onsite power generation, is a lignite occurring near Toledo, in Lewis County (2). The analysis of the raw coal follows:

	<u>As received</u>	<u>Moisture-free</u>
Moisture, percent	34.1	----
Volatile matter, percent	22.6	34.3
Fixed carbon, percent	19.3	29.3
Ash, percent	24.0	36.4
Sulfur, percent	.5	.7
B.t.u., per pound	5,070	7,700

When fresh the lignite is tough and resists degradation, but on drying it disintegrates practically to a powder. The bed contains numerous partings of clay; most are friable compared to the coal. Thus, as shown by the specific-gravity analyses in table II, the impurity tends to concentrate in the finer sizes. Material finer than

TABLE II. - Specific-gravity analyses of raw Toledo lignite

<u>Sizes, inches and mesh</u>	<u>Specific gravity</u>	<u>Weight-percent</u>	<u>Ash^{1/} percent</u>	<u>Cumulative</u>	
				<u>Weight-percent</u>	<u>Ash^{1/} percent</u>
1-1/2 to 20 Weight, 95.6 percent	Under 1.40	49.3	17.3	49.3	17.3
	1.40 to 1.50	17.0	32.2	66.3	21.1
	1.50 to 1.70	16.4	43.8	82.7	25.6
	Over 1.70	17.3	75.9	100.0	34.3
Under 20 Weight, 4.4 percent	Under 1.40	2.4	9.4	2.4	9.4
	1.40 to 1.50	13.6	18.8	16.0	17.4
	1.50 to 1.70	23.6	32.4	39.6	26.3
	Over 1.70	60.4	76.4	100.0	56.6
Composite, 1-1/2 to 0 Weight, 100.0 percent	Under 1.40	45.9	17.3	45.9	17.3
	1.40 to 1.50	16.4	31.7	62.3	21.1
	1.50 to 1.70	16.3	43.1	78.6	25.7
	Over 1.70	21.4	76.0	100.0	36.4

^{1/} Moisture-free basis

20 mesh contained 60 percent of sink 1.70, compared with 17 percent in the plus-20-mesh size.

A cleaning trial on this material in the Baum-type jig provided the results summarized in the following tabulation.

<u>Product</u>	<u>Weight-percent</u>	<u>Ash, percent</u>
Washed coal	75.1	25.1
Refuse	14.2	70.5
Slimes	10.7	70.5
Total	100.0	36.4

This clay did not swell enough to cause a troublesome decrease in density, but it disintegrated readily in the jig bed and became suspended in the water. Thus, 10.7 percent of the jig feed, nearly equaling the combined amount of draw and hutch refuse, left the system with the overflow of the washed-coal drag-settling tank.

Oregon Subbituminous Coal

The third coal investigated was from the Southport bed, Coos Bay field, Oregon. It is subbituminous B in rank and does not weather or slack as readily as

the two discussed previously. An analysis of the raw coal is shown in the following tabulation.

	<u>As received</u>	<u>Moisture-free</u>
Moisture, percent	16.2	----
Volatile matter, percent	29.2	34.8
Fixed carbon, percent	36.4	43.5
Ash, percent	18.2	21.7
Sulfur, percent	.8	.9
B.t.u., per pound	8,673	10,350

The coalbed contains a 7- to 8-inch parting of clay which swells readily, is rather friable, and therefore contaminates predominately the finer sizes of coal. As shown by the specific-gravity analyses in table III, the fraction finer than 20

TABLE III. - Specific-gravity analyses of raw Coos Bay coal

<u>Size, inches and mesh</u>	<u>Specific gravity</u>	<u>Weight-percent</u>	<u>Ash, ^{1/} percent</u>	<u>Cumulative</u>	
				<u>Weight-percent</u>	<u>Ash^{1/} percent</u>
2 to 20 Weight, 92.1 percent	Under 1.40	73.2	9.1	73.2	9.1
	1.40 to 1.50	10.4	20.6	83.6	10.5
	1.50 to 1.70	3.5	37.1	87.1	11.6
	Over 1.70	12.9	81.2	100.0	20.6
Under 20 Weight, 7.9 percent	Under 1.40	49.0	8.0	49.0	8.0
	1.40 to 1.50	11.0	18.4	60.0	9.9
	1.50 to 1.70	7.1	32.9	67.1	12.3
	Over 1.70	32.9	80.3	100.0	34.6
Composite, 2 to 0 Weight, 100.0 percent	Under 1.40	71.3	9.0	71.3	9.0
	1.40 to 1.50	10.4	20.4	81.7	10.5
	1.50 to 1.70	3.8	36.5	85.5	11.6
	Over 1.70	14.5	81.0	100.0	21.7

1/ Moisture-free basis

mesh contained 32.9 percent of heavy impurity, whereas the coarser size contained only 12.9 percent.

The behavior of this clay in the laboratory Baum-type jig differed from that of the other two described previously. When exposed to water, much of it disintegrated, but not to the fine particles that become suspended in the water. The disintegrated particles were of the size that reported predominately to the hutch product. As shown in the following tabulation, 5.8 percent of the feed, representing a substantial portion of the total clay, was removed as hutch refuse.

<u>Product</u>	<u>Weight-percent</u>	<u>Moisture-free ash, percent</u>
Washed coal	83.2	11.9
Draw refuse	7.6	68.7
Hutch refuse	5.8	56.9
Slimes	3.4	93.3

Another 3.4 percent of the feed was virtually pure clay suspended in the overflow of the washed-coal drag-settling tank.

This clay differed from the others in another respect also. More of it formed plastic masses into which particles of clean coal become imbedded. These aggregates of clay and coal were light enough to cause them to report to the clean-coal product, thus interfering with elimination of the clay.

Conclusions

Experience in cleaning only three low-rank coals affords little basis for generalization, because coals are inherently variable, each one presenting its own peculiar preparation problems. However, the similarity in the characteristics of the clays associated with these three coals suggests that these properties may be common to many of the clays associated with low-rank coals.

The pronounced tendency of these clays to disintegrate in water, to form plastic masses, and to swell with resulting decrease in density, have definite implications in terms of the design and operation of preparation plants. First, water clarification will be difficult and costly, involving extra filter capacity or settling ponds. Flocculation may be a prerequisite for acceptable filter capacity or adequate settling. Second, owing to a combination of particle size, stickiness, and swelling, this type of clay cannot be eliminated in washing as completely as the usual shales. Third, the jig offers certain advantages in dealing with clays that swell. The shearing, scrubbing action of the jig bed disintegrates and suspends in the wash water pieces of clay which otherwise would report to the clean product, leaving final elimination of the clay to a subsequent liquid-solid separation. In a sense, the jig acts as a combination log washer, classifier, and gravity-concentrating device.

Acknowledgement

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Literature Cited

- (1) Berkovitch, I., Manacherman, M., Potter, N. M., J. Inst. Fuel 32, 579-89 (1959).
- (2) Davis, C. W., Vacher, H. C., Conley, J. E., Bureau of Mines Tech. Paper 609, 15 (1940).
- (3) Yancey, H. F., Geer, M. R., Bureau of Mines Rept. of Investigations 3795 1-14, (1945).

DISTRIBUTION OF MACERALS IN THE PRODUCTS OF
VARIOUS COAL CLEANING PROCESSES

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Coal is comprised of several petrographic constituents, which are called macerals¹³ in analogy with the minerals in a rock. These macerals, differing from each other in their physical and chemical properties, govern the technological properties of coal. For example, in the case of coking coal, the carbon-rich "carbinite" macerals such as fusinite, semi-fusinite, and micrinite are inert on heating, and consequently reduce the caking index. The hydrogen-rich "hydrinite" macerals like exinite, resinite, and alginite decompose on heating into plastic melt and tar. In contrast, the maceral vitrinite constitutes the actual coking principle.

For the sake of more efficient utilization, numerous attempts have been made in research laboratories for separating coal macerals by means of controlled crushing and sizing^{1,5,8,9}, heavy liquid separation^{2,4,10}, and flotation^{6,7,11}, but no similar study has been made in commercial coal preparation plants. The object of this work was to determine the effects of various concentration methods on the distribution of macerals in the products of two Pennsylvania bituminous coal cleaning plants. Also determined were the sulfur and ash contents of the fractionized maceral concentrates.

Experimental.

Each coal sample was first crushed and pulverized to minus 100-mesh and then ground to minus 10-micron. About 5 grams of the ground sample was mixed with 200 ml. of an aqueous zinc chloride solution in a glass bottle, and agitated for 14 hours. The resulting suspension was separated into five different "maceral concentrates" according to the float-and-sink method of Figure 1. A centrifuge was used to aid the separation. The concentrates thus obtained were washed, dried, weighed, analyzed chemically for their ash and sulfur content, and examined microscopically for their petrographic composition.

The procedure of microscopic examination consisted of mounting each maceral concentrate with paraplex. The mounted sample was polished, and then examined under a Leitz, Panphot microscope with 750X resultant magnification and arc illumination. A series of traverses, one millimeter apart, were completed on each block until a total of 300 entities had been identified and counted. The results, as given in Table I, were used as a basis for calculating the distribution of macerals in the products of various coal cleaning processes.

Results and Discussion.

Figure 2 shows that the percentage recovery of macerals in the two Pennsylvania coal cleaning plants decreases in the order of exinite, vitrinite, micrinite, fusinite, and mineral matter, irrespective of the employed concentration method. This is caused, in the case of gravity concentration, by the increase of the density of the macerals in the same order. The reason for the different floatabilities of the macerals lies in the variation of their chemical composition, as indicated partly in Figure 3. A detailed explanation has been given in a previous publication.¹²

Figure 2 shows also that the relative effectiveness of the various concentration methods for separating the desired macerals vitrinite, exinite, and micrinite from the undesired maceral fusinite and mineral matter decreases generally in the order of heavy media, Rheclaveur, tabling, and flotation. This is due, as shown in Table 2, chiefly to the decrease of the particle size of coals treated by these processes in the same order. Fine coal particles, having a tendency of indiscriminate flocculation, are more difficult to clean than coarse ones. Comparing with particle size, the influence exerted by the petrographic composition of the tested coals is relatively insignificant, because of limited variation. The summation curves³ show that vitrinite is more selectively upgraded by all the tested concentration methods than the rest of macerals.

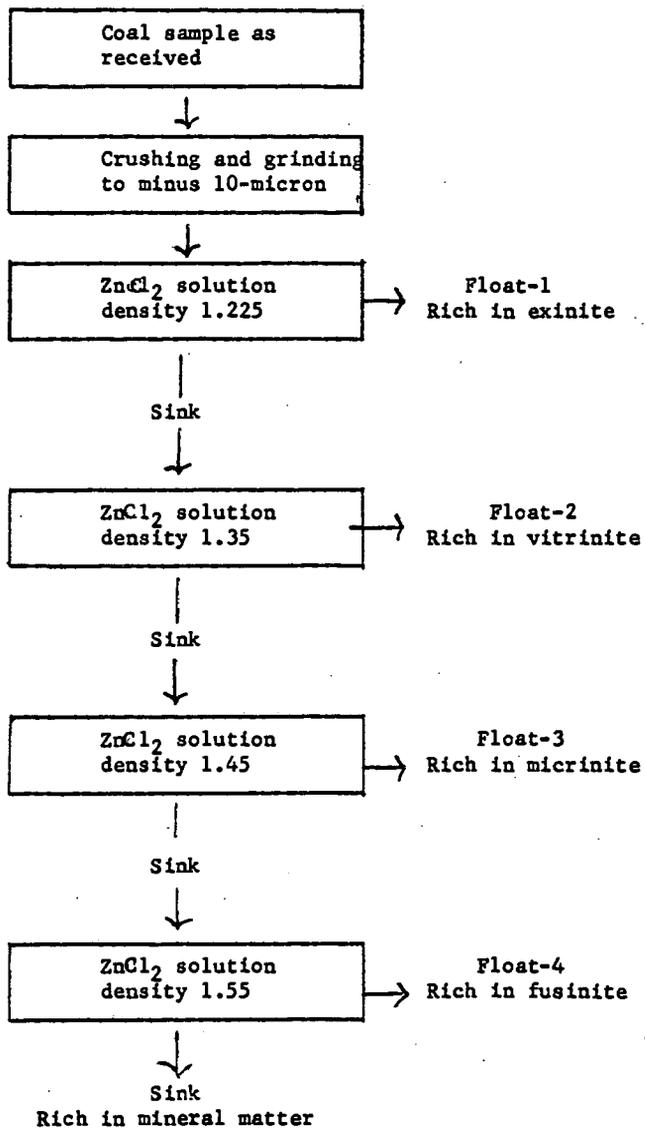


Figure 1. The Float-and-Sink Method Used for Fractionizing Coal Macerals.

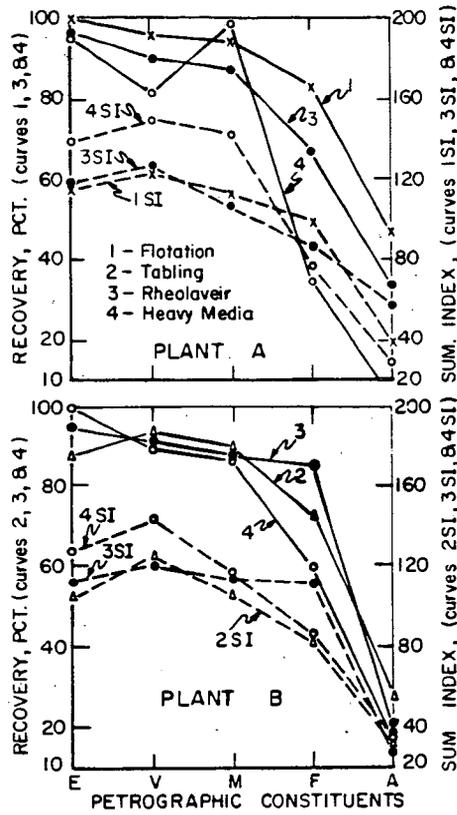


Figure 2. The Upgrading of Macerals With Various Concentration Methods in Two Pennsylvania Bituminous Coal Cleaning Plants.

Petrographic constituents: E, exinite; V, vitrinite; M, micrinite; F, fusinite; A, mineral matter.

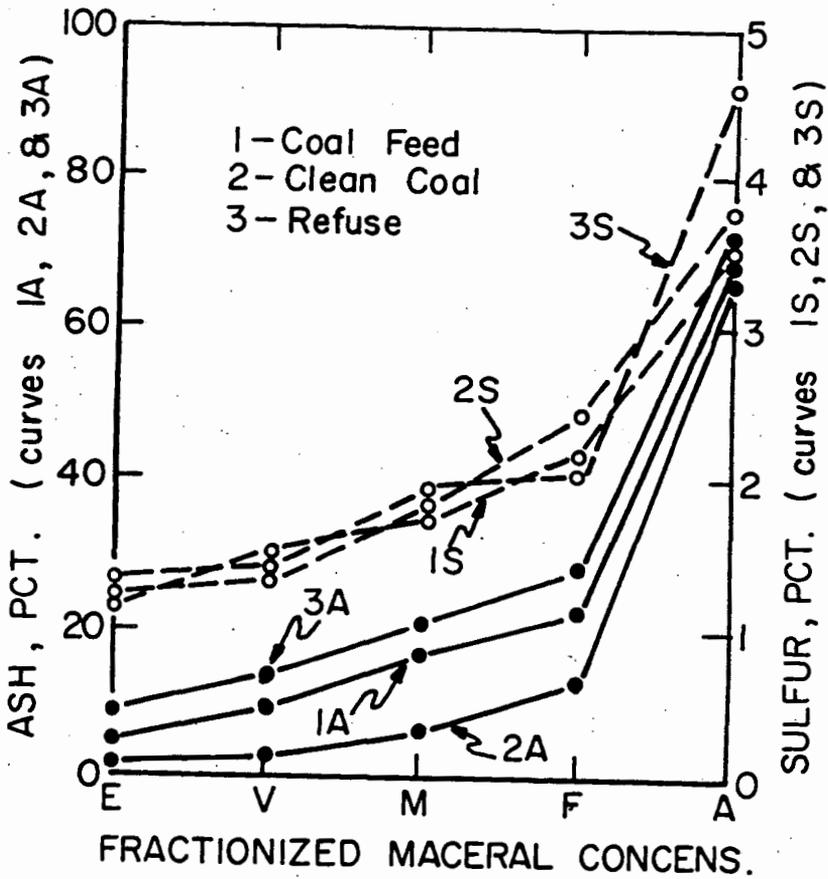


Figure 3. The Ash and Sulfur Content of Fractionized Coal Maceral Concentrates.

Table 1. The Petrographic Composition of Fractionized Maceral Concentrates, as Determined by Microscopic Examination.

Fractionized maceral concentrate	Volume Percentage of petrographic constituent						Intended constituent
	Exinite	Resinite	Vitrinite	Micrinite	Fusinite	Mineral matter	
Exinite	5	5	67	4	18	1	10*
Vitrinite	1	10	79	6	3	1	79
Micrinite	4	1	72	12	10	1	12
Mineral matter	Trace	Trace	16	2	12	70	70

* Including resinite.

Table 2. The Particle Size and Petrographic Composition of Feeds for the Various Concentration Processes of Two Pennsylvania Coal Cleaning Plants. (Volume %)

Particle size and petrographic composition of feed	Plant A			Plant B		
	Heavy media	Rheola- veur	Flotation	Heavy media	Rheola- veur	Tabling
Particle size	3/8" x 6"	3/8" x 0	-48 mesh	1/4" x 8"	1/4" x 0	1/8" x 0
Exinite	6.26	7.50	5.30	6.95	8.59	9.57
Vitrinite	53.60	64.07	60.52	57.56	62.18	65.61
Micrinite	4.20	5.72	6.64	4.79	4.22	4.99
Fusinite	9.81	8.38	11.21	8.17	14.31	7.65
Mineral matter	26.21	14.34	16.36	22.52	10.69	12.22

Literature cited

1. Anon., The Petrographic Preparation of Coals for Coking, Coke and Gas, pp. 246-256, (July 1956).
2. Alpern, B., Proprietes Physico-Chimiques et Cokeifiantes des Macerats de Quelques Charbons en Fonction de Leur Degre de Houillification, Proceedings of the International Committee for Coal Petrology, (Liege, Belgium), No. 2, pp. 43-46, (1956).
3. Gillies, G. A., Lyle, A. G., and Dunkle, J. D., A Graphical Method for Evaluating Selective Flotation Tests, Technical Publication 1409, Mining Technology, AIME, Vol. 6, pp. 1-12, (February 1942).
4. Horton, L., Separation of Coals Into Fractions of Different Densities, Fuel, Vol. 31, pp. 341-354, (1952).
5. Lehmann, K., and Hoffman, E., Coal Dressing from Petrological Point of View, Gluckauf, Vol. 67, pp. 1-14, (1931).
6. Maier, L. M., Tzukanian, L. E., and Luisenl, P. D., Estimation of Fusain from Coal by Means of Flotation, Coal and Chem., (USSR), No. 5-6, pp. 104-112, (1934).
7. Price, F. C., Treatment of Coal, U. S. Patent 1,499,872, (July 1, 1924).
8. Stevens, J. L., The Petrographic Treatment of Coal, Colliery Guardian, Vol. 142, pp. 572-6 and 667-8, (1931).
9. Sasaki, Noburo, Improvement of Coking Ability by Petrographical Grinding of Coals, Journal of Coal Research Institute (Japan), Vol. 6, pp. 321-9, (1955).
10. Sasaki, Noburo, Petrographical Preparation of Coals, Journal of Coal Research Institute, (Japan), Vol. 6, pp. 400-6, (1955).
11. Sasaki, Noburo, Distribution of Petrological Constituents of Coal Prepared by Flotation, Journal of Coal Research Institute, (Japan), Vol. 1 pp. 67-73, (1950).
12. Sun, S. C., Hypothesis for Different Floatabilities of Coals, Carbons, and Hydrocarbon Minerals, Trans. AIME, Vol. 199, pp. 67-75, (1954).
13. Van Krevelen, D. W., and Schuyer, J., Coal Science, pp. 50-75, (1957), Elsevier Publishing Co., New York

FACTORS IN EVALUATION OF NEW METHODS OF LIBERATION AND SEPARATION
OF PYRITE AND OTHER MINERALS FROM COAL

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Introduction

Present methods of coal cleaning are based on one or more of these common factors:

- (1) appearance - as in hand cleaning
- (2) shape and toughness - as in Bradford-type rotary breaker
- (3) specific gravity - as in pneumatic cleaning, jigging, and heavy media separation
- (4) surface phenomena - as in froth-flotation or oil agglomeration

Electrostatic separators are widely used in mineral beneficiation, especially for beach sands, but have not achieved much acceptance in the coal industry.

Before considering factors involved in new methods of liberation and separation of mineral matter from coal, the limitations of the methods already in use should be recognized. These limitations are mainly economic but some are based upon a distinct lack of appreciation for the mode of occurrence of mineral matter in coal. Large sizes of coal can be economically cleaned by simple gravity methods and suitable products are prepared for the market by crushing and screening the cleaned coal. Fine coal is either (a) cleaned separately and blended with larger sizes, (b) blended uncleaned with larger sizes, (c) rarely cleaned and sold as a separate product, or (d) discarded. With ever increasing application of mechanical mining techniques the larger proportion of fines produced now prohibits any coal to be discarded other than extreme fines, and even these fines, because of water clarification problems, are being recovered at some preparation plants.

There is no problem in beneficiation where mineral matter in the form of large pieces of roof and floor rock, thick partings, large concretions or lenses of clay, shale, and pyrite are the common impurities. Ordinary jigging or the use of a sand cone will readily remove these gross impurities from the coal. Separation and removal of large, tough, tabular pieces of rock and mineral is simply performed by the Bradford rotary breaker. The details of these devices and almost every other coal preparation apparatus are given in "Coal Preparation," edited by D. R. Mitchell.⁽¹⁾

Fine coal cleaning may be considered from two different viewpoints, depending on the nature of the coal and its associated impurities and depending on the ultimate market. Where a low grade coal is being processed the fine coal may not require cleaning prior to blending with other coal unless the percentage of non-combustibles is so high as to severely affect the quality of the entire blend. For premium grade coal, especially metallurgical coal, the fines may represent an appreciable percentage of low-sulfur coal - an irreplaceable commodity.

Fine coals, down to about 48 mesh, may be cleaned on concentrating tables, or by froth flotation, a process which is restricted almost exclusively to the pre-

mium low-sulfur coals. With a sufficient number of cells it appears that there is no lower limit to the size of the coal cleaned by froth flotation. Two methods of washing fine coal that have recently been introduced from abroad are modifications and extensions of the equipment already used in coal preparation plants - the feldspar jig⁽²⁾ and the Dutch State Mines heavy medium cyclone.⁽³⁾

Some concept of the magnitude of fine coal cleaning is to be gained from inspecting the data in Table I which shows the tonnages and percentages of bituminous coals and lignite mechanically cleaned in the years 1948 and 1958.

TABLE I. COMPARATIVE DATA FOR BITUMINOUS COALS AND LIGNITES MECHANICALLY CLEANED DURING 1958 AND 1948⁽⁴⁾

	1000's of Net Tons		Percentage of Total	
	1958	1948	1958	1948
Jigs	115,321	87,500	44.5	48.4
Concentration Tables	18,142	4,360	7.0	2.4
Classifiers	8,793	18,304	3.4	10.1
Launders	6,768	16,768	2.6	9.3
Dense Medium Processes	52,735	20,638	20.4	11.4
Jigs and Tables	10,076	5,252	3.9	2.9
Other Combinations	28,318	11,816	10.9	6.5
Pneumatic	18,882	16,216	7.3	9.0
Total	259,035	180,880	100.0	100.0
Percentage of Total Coal				
Production Mechanically Cleaned	63.1	30.2		
Percentage of Refuse Discarded From Raw Coal	19.3	16.0		

These figures, taken from the Bureau of Mines Yearbook for 1958⁽⁴⁾ are the latest such complete data publicly available. The twofold increase in the percentage of coal mechanically washed from 1948 to 1958 is not at all surprising since the rate of mechanical mining and machine loading has sharply increased during the same time interval. Consequently, fine coal cleaning has become increasingly important, and statistics on fine coal cleaning facilities show an upward trend in the application of the older, established methods of fine coal cleaning. Acceptance of the newer techniques of fine coal cleaning, such as application of the feldspar jig, heavy media cyclones, and the recently repopularized froth flotation of coal fines has not yet reached the level that significant trends can be evaluated - but there can be no doubt that there will be an increase in the use of these techniques. Data from "Coal Age"⁽⁵⁾ show that during 1959 seven froth flotation plants with a combined capacity of 313 tons per hour were contracted for by the coal industry, while "Coal Age"⁽⁶⁾ shows that during 1958 eight froth flotation plants were contracted for, of which six had a combined capacity of 149 tons per hour with the capacities of the other 2 plants not disclosed. A capacity of 462 tons per hour may not seem impressive, but the aggregate capacity of these new plants alone, operating at two shifts a day for only 200 days, becomes more than 180,000 tons a year.

The application of these techniques to fine coal cleaning has resulted in a decrease in the waste of a valuable raw material, and with proper care, at no reduction in the quality of the coal shipped to the consumer.

New Factors

All commercial processes for cleaning coal are based upon practical application of standard washability tests. Generally these sink and float washability tests are adequate for coal preparation plants which are designed to meet specifications based on these tests. Factors that have not been considered in the design of coal cleaning plants are:

- (1) Size distribution of mineral particles in the coal.
- (2) Spatial arrangement of minerals through the coal substance.
- (3) Differential grindability of coal and its constituents, and the influence of mineral matter composition on coal grindability.

Size of Mineral Particles

The size of mineral particles and their distribution is fundamental to the washability characteristics of any coal. In making routine washability tests at any predetermined specific gravity the sole controlling factor as to whether a free-settling particle will report to the sink or to the float fraction is the proportion of high-specific-gravity mineral matter in that particle. Where a large fragment sinks because a small single grain of mineral matter is included, it is a relatively simple matter to recycle that particle to a crusher and to make the separation on a second pass. But where a similar large particle contains an equivalent amount of mineral matter as many small disseminated grains, rather than as one large grain, simple crushing will fail to release sufficient mineral particles from the coal to permit a clean separation by gravity methods. Fortunately, this has not been the situation for many of the commercially important coals, otherwise beneficiation by the relatively simple means heretofore developed would not have been possible. The issue now is to what extent does this condition prevail in coals which cannot be beneficiated to a more desirable level of ash and sulfur content by conventional coal cleaning methods?

An estimation of the relative size of mineral grains in coal can be made from grain counts and size measurements of minerals in surface sections of coal by micrometric methods. In the example to be cited only measurements of pyrite grains were made because of the emphasis on sulfur in the project for which the research was conducted. Polished surface sections were made of samples of five test coals and of separates prepared from these coals as described here:

- (1) raw run-of-mine coal crushed to 1/2" x 0"
- (2) float fraction (1.58 Sp. Gr.) of 30 mesh x 0" coal
- (3) sink fraction (1.58 Sp. Gr.) of 30 mesh x 0" coal
- (4) float fraction (1.58 Sp. Gr.) of 1/2" x 30 mesh coal

Micrometer measurements were made on at least 500 to 1000 pyrite grains from samples prepared from each of the separates described for all five test coals. These data are summarized in Table II. While quantitative as to the size of pyrite grains relative to each other, the data do not take into account the grouping or bunching of pyrite grains at any particular locus or along a particular plane.

From the data in Table II it can be seen that a very large percentage of pyrite particles are extremely small, with from 70 to 98 percent of the measured particles smaller than 20 microns in diameter. Yet, with such a range of sizes, the relative weight percentage of these particles ranges from a low of about 1.3

TABLE II. PYRITE PARTICLE SIZE DATA FOR TEST COALS AND THEIR FLOAT-SINK FRACTIONS (Sp. Gr. 1.58)

(Data are reported as the percentage of pyrite particles within a size range and as the relative weight percent of particles as determined by computing the average volume of pyrite particles within the size range.)

Size range, microns	Average vol. of particles, cu. microns	Original Coal		Float		Sink		Float	
		1/2" x 0" % Particles in size range	Relative wt. %	30 mesh x 0" % Particles in size range	Relative wt. %	30 mesh x 0" % Particles in size range	Relative wt. %	1/2" x 30 mesh % Particles in size range	Relative wt. %
Kentucky No. 11 Coal									
< 5	15.6								
5 - 20	1950	40.0	0.2	66.8	0.5	58.7	--	26.8	--
20 - 40	27 x 10 ³	55.0	42.5	51.6	33.0	58.7	0.5	65.4	11.3
40 - 75	18.9 x 10 ⁴	4.9	49.7	1.1	16.0	23.2	2.7	7.4	18.3
75 - 150	19.5 x 10 ⁵	0.1	7.6	0.5	50.5	11.6	9.4	--	--
150 - 250	8 x 10 ⁶	--	--	--	--	5.1	42.0	0.4	70.4
> 250	52.5 x 10 ⁶	--	--	--	--	1.2	41.0	--	--
						0.2	4.4	--	--
Pittsburgh Bed Coal (W. Va.)									
< 5	15.6								
5 - 20	1950	25.7	--	44.8	0.1	48.9	--	45.2	--
20 - 40	27 x 10 ³	44.8	1.3	44.0	13.9	48.9	0.2	37.8	3.0
40 - 75	18.9 x 10 ⁴	17.3	7.0	9.8	43.0	29.2	1.5	13.0	14.6
75 - 150	19.5 x 10 ⁵	2.2	2.8	1.4	43.0	11.5	0.4	3.3	25.8
150 - 250	8 x 10 ⁶	0.2	64.7	--	--	7.5	27.7	0.7	56.6
> 250	52.5 x 10 ⁶	--	24.2	--	--	2.5	30.4	--	--
						0.4	39.8	--	--
Meigs Creek No. 9 Coal (Ohio)									
< 5	15.6								
5 - 20	1950	31.5	0.1	28.7	--	38.4	--	36.9	0.1
20 - 40	27 x 10 ³	60.5	13.5	59.4	13.0	38.4	0.3	54.5	17.8
40 - 75	18.9 x 10 ⁴	6.8	20.9	8.8	27.0	50.4	3.3	7.0	31.6
75 - 150	19.5 x 10 ⁵	1.0	21.4	3.1	60.0	15.4	11.8	1.6	50.5
150 - 250	8 x 10 ⁶	0.2	44.3	--	--	4.2	33.0	--	--
> 250	52.5 x 10 ⁶	--	--	--	--	1.6	51.6	--	--
Illinois No. 6 Coal									
< 5	15.6								
5 - 20	1950	54.2	0.2	54.6	0.1	35.7	--	52.3	0.1
20 - 40	27 x 10 ³	36.1	1.6	38.0	9.0	41.0	0.5	42.9	14.6
40 - 75	18.9 x 10 ⁴	6.7	2.4	6.2	20.4	14.7	2.3	2.6	12.3
75 - 150	19.5 x 10 ⁵	2.0	8.6	1.0	23.0	5.6	6.2	2.2	73.0
150 - 250	8 x 10 ⁶	0.7	32.4	0.2	47.5	1.4	16.0	--	--
> 250	52.5 x 10 ⁶	0.3	54.8	--	--	1.6	75.0	--	--
Tobo Coal (Missouri)									
< 5	15.6								
5 - 20	1950	81.5	0.1	70.0	0.2	34.2	--	63.3	0.1
20 - 40	27 x 10 ³	16.1	2.4	23.9	6.7	42.1	0.8	25.5	2.7
40 - 75	18.9 x 10 ⁴	1.9	3.9	4.2	16.3	10.3	2.6	8.4	12.2
75 - 150	19.5 x 10 ⁵	0.2	2.8	1.8	48.8	8.2	1.5	2.2	22.3
150 - 250	8 x 10 ⁶	0.1	29.8	0.1	28.0	5.2	95.1	0.6	62.7
> 250	52.5 x 10 ⁶	--	61.0	--	--	--	--	--	--

weight percent to a high of about 42.7 percent. This means that, except for the Kentucky No. 11 coal, the amount of pyritic sulfur in pyrite grains less than 20 microns in size is negligible. In contrast, for three of the coals, most of the pyrite is in grains larger than 75 microns (plus 200 mesh). This information can be used in two ways: to show that dissemination of fine grained minerals throughout a coal could, if no moderating factors are involved, completely abrogate established coal cleaning practice; and, to show to what extent these moderating factors have influenced coal cleaning. Knowing this, it may then become possible to apply this information to improvement of coal cleaning processes and to the beneficiation of coals, which, despite inherently desirable properties and characteristics, contain too much fine grained pyrite and other minerals to permit their utilization in the premium markets.

Arrangement of Minerals in Coal

Table III shows what might be expected from certain coal-pyrite mixtures if all the pyrite were uniformly disseminated. Such expectations would be most disheartening to any preparation plant manager. For example, with two percent by volume of pyrite, all the sample would sink at specific gravity 1.25, and all would float at specific gravity 1.30.

TABLE III. SPECIFIC GRAVITY FACTORS FOR MIXTURES OF PYRITE AND COAL

<u>Vol. % Pyrite</u>	<u>Wt. % Pyrite</u>	<u>Wt. % Pyritic Sulfur</u>	<u>Calculated Specific Gravity of Mixture</u>
1	4.04	2.16	1.238
2	7.84	4.18	1.276
4	14.8	7.9	1.352

Assume: Sp. Gr. Mineral-free Coal = 1.2, Sp. Gr. Pyrite = 5.0,
Sp. Gr. Non-Pyritic Minerals = 2.5

Table IV is a somewhat more realistic array of figures showing what coal producers would be faced with if all the mineral matter in coal was present in a highly disseminated form. The ash and pyritic sulfur figures in Table IV are equivalent to that of many fine, high grade premium coals mined today. But imagine what a dismal future any producer would have if a coal with 10 percent ash and almost two percent pyritic sulfur could not be greatly beneficiated by washing at a 1.30 specific gravity?

To know that the washability of coals is dependent upon the amount and distribution of minerals in coal is not enough. What is necessary is an understanding and appreciation of how the minerals occur in every hard-to-wash coal of potential value. Research on methods to beneficiate these coals, though not immediately successful, must result in techniques that would enhance the washability characteristics of all other coals.

Microscopic examination of polished surface sections of coal shows that the minerals in coal, and especially pyrite, are largely present in some regularized pattern. Despite the seeming randomness of occurrence of pyrite and other minerals, low power magnification shows individual pyrite grains to be concentrated in planar distributions which are roughly, and often ideally, parallel to the bedding plane of the coal. This is illustrated in Figures 1, 2 and 3. Pyrite, which is so clearly recognized in photomicrographs taken with reflected light, is representative of the

occurrence of other minerals which are interbedded with the coal, but which may have a different origin. (7)

TABLE IV. SPECIFIC GRAVITY, PYRITIC SULFUR AND ASH DATA FOR MIXTURES OF MINERAL MATTER, PYRITE AND COAL

Non-Pyritic Minerals		Pyrite		Pyritic Sulfur	*Theoretical Ash	Sp. Gr.
Vol. %	Wt. %	Vol. %	Wt. %	Wt. %	Wt. %	
1	2.6	--	--	--	3.1	1.213
1	1.9	1	4.0	2.14	4.3	1.251
2	3.95	1	3.95	2.12	7.4	1.264
4	7.75	1	3.87	1.94	10.1	1.290
2	4.02	0.5	2.01	1.09	6.0	1.245

Assume: Sp. Gr. Mineral-free Coal = 1.2, Sp. Gr. Pyrite = 5.0, Sp. Gr. Non-Pyritic Minerals = 2.5

*% Theoretical Ash \approx 0.5 + Wt. % Non-Pyritic Minerals + Wt. % Pyrite (0.73)

The ready separation of coal from mineral partings within the coal bed is an example of the kind of bonding weakness that commonly exists between the layers of material which comprise a complete coal bed. Upon handling and primary crushing further breakage takes place along similar planes. When coal is crushed to even finer sizes by some differential method, to take full advantage of the planar arrangement of minerals on a microscopic scale, it will then be possible to design economical cleaning plants for fine coal. In such a plant coal will be reduced to fine sizes according to predetermined procedures based on mineral relationships established for that coal. Subsequent separation will depend upon the market for which the coal is being prepared.

Limitations in Application of New Factors

One of the principal reasons these new factors have not been given serious consideration in the solution of coal cleaning problems is that very little is known about the minerals in coal other than that they are undesirable impurities. Another limitation may perhaps be related to the first: there is a lack of qualified personnel now engaged in research on these problems. Investigations of the nature of the minerals in coal and of the variations in the mineral matter content of coal beds on a systematic basis should have been supported for many years.

There has been too much concern with sudden changes in bed characteristics which may result in increased ash content or increased sulfur content of prepared coals, and not enough interest and concern for the careful accumulation of data on minerals in coal which might indicate why these variations in mineral matter content and sulfur content do occur. Before new methods of liberation and separation of minerals from coal can be applied on a wide scale, a great deal of data must first be developed so that engineering can be applied most effectively.

As in any other widespread modern industry, standards and standard tests provide a common basis for exchange of information and intelligence for the coal industry. Rigid or nearly rigid adherence to standard tests is a commendable practice. And this may be especially true if there has been a battle over the acceptance or modification of the standards. Yet it appears that for many tests the data are merely recorded, collated, reported, and subsequently filed for reference. If the tests are at all worth-while doing then the data from these tests should be evaluated

and re-evaluated so that further testing or re-analysis of the data may have greater significance. One such test is the Hardgrove grindability test. Recent testing of the Hardgrove grindability method indicates that mineral matter is an important factor in determining the Grindability Index, and that perhaps high mineral matter content coals (high ash coals) are given Indexes that are too high.

Recommendations

A discussion of significant factors followed by a discussion of the limitations in the application of these factors would be valueless without a few recommendations as to what should be done. These are a few recommendations:

(1) New methods of evaluation of coal cleaning methods should be based on tests other than simple sink and float.

(2) Size reduction methods must be developed for coal which will permit maximum liberation of mineral matter, while at the same time limiting the amount of extreme fines produced.

(3) Sieving fine coal has been costly, but newly designed sieving machines are now being offered which will permit close sizing with a high throughput at relatively low cost. Air cleaning of such closely sized coal should be investigated.

(4) Although many coal cleaning machines now on the market have established certain lower size limits for feed material, it is possible that where good liberation of pyrite and other minerals can be assured this lower limit can be pushed yet lower. For example, the Dutch State Mines heavy media cyclone will treat fine coals down to 48 mesh or at best 60 mesh. At such sizes satisfactory products have been prepared, but the limit could be pushed to 100 mesh or possibly even to 140 mesh if the prime consideration was to separate coal, with specific gravity of about 1.30, from pyrite which has a specific gravity of 5.0.

Acknowledgment

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REFERENCES

- (1) Mitchell, David R., "Coal Preparation," A.I.M.E., New York (1950)
- (2) Geer, M. R., and Yancey, H. F., "Operating Results With the Feldspar Fine-Coal Jig," U. S. Bureau of Mines R. I. 5412 (1958)
- (3) Krijgsman, C., "The Dutch State Mines Heavy Medium Cyclone Washing System," Parts I and II, Mining Congress Journal, 45, pp. 47 and 91 (1959)
- (4) U. S. Bureau of Mines Minerals Yearbook 1958, p. 100 (1959)
- (5) "Mining, Stripping, Preparation in 1959," Coal Age, p. 80, Feb. 1960
- (6) "Mining, Stripping, Preparation in 1958," Coal Age, p. 80, Feb. 1959
- (7) Deul, Maurice, "The Principal Geological, Chemical and Physical Factors Controlling the Mineral Content of Coal," Preprints of papers presented at Boston Mass., American Chem. Soc., Div. of Gas and Fuel Chem., Vol. II, p. 21 (1959)

Figure 1. Euhedral pyrite crystals
in Meigs Creek No. 9 coal from
Harrison County, Ohio.

————— = 40 Microns



Figure 2. Disseminated pyrite in
Meigs Creek No. 9 coal from
Harrison County, Ohio.

————— = 40 Microns

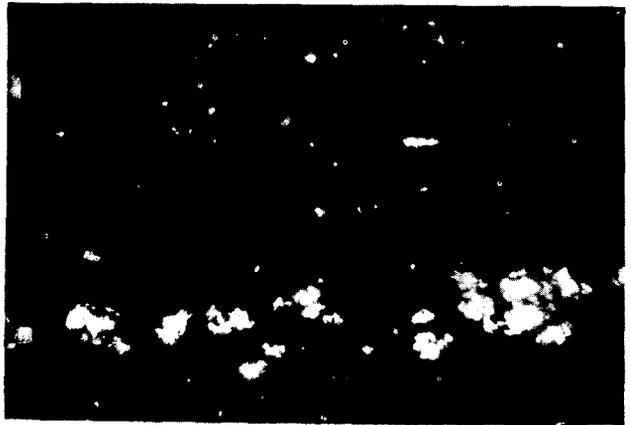
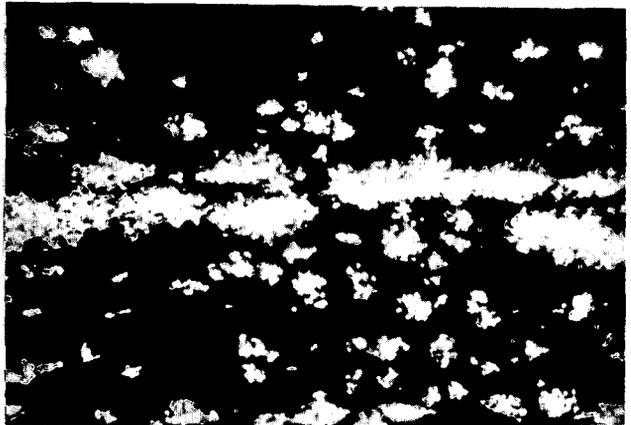


Figure 3. Disseminated pyrite in
Meigs Creek No. 9 coal from
Harrison County, Ohio.

————— = 40 Microns



SOME PRACTICAL CONSIDERATIONS FOR THE APPLICATION
OF COAL PULVERIZATION AT COKE PLANTS

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During recent years, the United States Steel Corporation has focused increased attention on coal-pulverization practice at coke plants.^{1-5)*} Coal pulverization at coke plants offers two opportunities for steel-plant cost improvement. First, coal blends that are known to give cokes of good strength may yield, through pulverization, additional increases in strength, such increases resulting in increased hot-metal production at the blast furnace. Second, by pulverization of poorer coking coals the amounts of the more costly and less readily available blending coals may be reduced without a loss of coke strength.

Prior to any discussion of coal pulverization, it is important to realize that an optimum pulverization level (commonly reported in terms of % minus 1/8-inch coal) exists for each coal or coal blend and that overpulverization can be detrimental.^{1,2,5-7)} Optimum pulverization for a given coal or coal blend generally will give some loss of bulk density (or coke production) with a corresponding increase in coke quality (or strength). Except with dry coals, however, overpulverization results in appreciable loss of bulk density and may reduce or give little improvement in coke strength, particularly the Hardness Factor, for normally coking coals. This loss of bulk density can be substantially recovered by adjusting the moisture content or by appropriate oil addition.⁸⁻¹⁰⁾ Also, means for increasing the drop velocity of the coal into the coke ovens are helpful. For poor coking coals, increased coking rates are useful. The primary objective in pulverizing coals is the improvement of coke quality to give more intimate contact between the coal constituents. Thus, pulverization permits homogenous mixing and intimate contact between the various constituents.

Once the need for a coke-plant coal-pulverization system has been established and the optimum pulverization level has been determined by carbonization tests, the next objective should be to attain in a practicable manner the required level of pulverization. However, this should be achieved so as not to exceed the minimum allowable increase of fine "bug dust" (arbitrarily defined as minus 100-mesh coal) over that level present in the unpulverized coals. Attainment of this objective will minimize the amount of bug dust in the coal and the loss in bulk density. Minimizing the amount of bug dust minimizes the loss of coal during pulverization and during carbonization, with attendant improvement in plant housekeeping and safety.

Coke-plant coal-pulverization systems fall into three basic types, as illustrated in Figure 1. These are (1) single-pass systems in which all coal to be pulverized is passed only through pulverizer mills, (2) prescreening systems in which the fine (commonly, minus 1/8-inch) coal is recovered by screening and in which the screen overproduct is pulverized and mixed with the recovered screen underproduct, and (3) closed-circuit systems in which screens are used before and after pulverizers to contain all oversize coal for repulverization until the coal is fine enough to be released from the system as screen underproduct.

As these systems increase in complexity, there is a corresponding improvement in the control of pulverization level. Table I illustrates the increased pulverization that can be achieved for a given coal by progressing from the single-

* See References.

pass to the prescreening to the closed-circuit systems. However, if the single-pass system were used to bring the level of pulverization at 1/8 inch to 86 per cent, the amount of bug dust would have been considerably greater than the 12.3 per cent attained with the prescreening system. Also, by using the more elaborate closed-circuit system, even greater improvements in the level of pulverization and in the control of bug dust can be realized.

In pulverizing coals for blending, the low-, medium- and high-volatile coals attain an optimum pulverization level at widely varying pulverizer-mill speeds. These mill speeds range from about 4500 feet per minute (fpm) rotor-tip velocity for certain soft and friable low-volatile coals to about 8000 fpm rotor-tip velocity for hard high-volatile coals. These mill-speed ranges were established by using impact-type coal pulverizers instead of hammermills, since the former lend themselves more readily to speed variation and give a more predictable and significant change in pulverization level for a change in mill speed. Also, for a given pulverization level, impact-type pulverizers will produce less bug dust than hammermills.

It is a popular, but perhaps obsolete, coke-plant practice to pulverize all coals by using hammermills (or hammermills with grates removed) operating at the same speed. The above conclusions, together with experience at coke plants, support the observation that operating pulverizer mills at a single speed will overpulverize the softer blending coals. Normally, the single-speed operation is suited for pulverizing high-volatile coals, but because of the high mill speeds used, overpulverization of the low-volatile coals results in, and is largely responsible for, excess production of bug dust.

The detrimental effects of pulverizing a softer and a harder coal at the same mill speed (6300 fpm rotor-tip velocity) are shown in Table II. High-volatile coal was satisfactorily pulverized, but low-volatile coal was overpulverized. The amount of bug dust in the low-volatile coal was almost double that in the high-volatile pulverized product. The improved pulverization that can be achieved by pulverizing softer coals at lower mill speeds (4400 fpm) and harder coals at higher mill speeds (6300 fpm) is shown in Table III. High- and low-volatile coals were pulverized to give the same amounts of bug dust.

For most American coke plants it appears that the use of a prescreening system incorporating impact pulverizers at two mill speeds is worthy of consideration. In large pulverization installations, fixed-speed mills (or direct-drive mills) may be used provided that a lower and a higher mill-speed section is available for handling the various coals. For smaller installations where the same series of mills must be used intermittently to pulverize the different coals to be blended, variable-speed drives are recommended. This permits preselection of optimum mill speed prior to pulverization of the individual low-, medium- or high-volatile coal.

Because of its merit, the combination of prescreening with two mill speeds in coke-plant coal pulverization has been adopted by the United States Steel Corporation and is being applied at the Corporation's large, new coal-pulverization facility at Gary Steel Works, Gary, Indiana.

In coke plants where pulverization is applied, but where a complete change from the older single-pass systems is not justified, it may be desirable to equip the mills with impactor-conversion assemblies and variable-speed drives. This practice can reduce the bug-dust level and improve control of pulverization.

Generally, with these older fixed-speed, single-pass pulverization systems, control of pulverization is sought by changing the clearance between the mill rotor and impact surface. However, tests indicate that feed rate may offer a more attractive means for pulverization control. It appears that for each mill studied, increased pulverization will be accompanied by increased bug dust, but beyond a certain mill feed rate (regardless of mill speed or setting) both pulverization and

bug dust decrease. The "reversal" of this trend is no doubt the result of portions of coal going through the mill unpulverized. The feed rate at which this reversal occurs (reversal-point feed rate) is at or near the manufacturer's so-called nominal rated mill capacity. Tests indicate that feeding a mill at a rate that exceeds the reversal point and approaches the mill stall point will result in an approximately equivalent percentage reduction in both the pulverization and bug-dust levels. (The stall point of most pulverizers occurs from 50 to 100 per cent above their nominal rated capacities, and at a number of plants, mills are being run at these levels.) Therefore, the attainment of a feed rate between the reversal point and the stall point, obtained by choke feeding, offers a means for reducing the amount of bug dust and increasing mill productivity, but is achieved with some reduction in level of pulverization. This reduction is not great and the new level of pulverization may be consistent with requirements. Table IV illustrates the effect on size-consist when the pulverization reversal point is exceeded.

Conclusions

From the data presented above, it may be concluded that the most effective method of using pulverizers in a single-pass system is to design the machine rotor speed to exceed that necessary to produce the desired pulverization level and then choke-feed the machine to depress pulverization back to the desired level. This practice will give both desired pulverization and significant bug-dust reduction. In those coke plants where the older fixed-speed, single-pass mills are in use, hard, high-volatile coals can be pulverized to a greater degree by feeding at a rate approaching the nominal rated capacity of the mill. Excess bug dust resulting from the soft, low-volatile coals can be reduced by increasing feed rate above the rated capacity of the mill to a value approaching the mill stall point. Therefore, by manipulating the feed rates of these older systems, pulverization and bug-dust levels of the different coals can, to a limited degree, be made to approach more closely the desired value for any given mill setting.

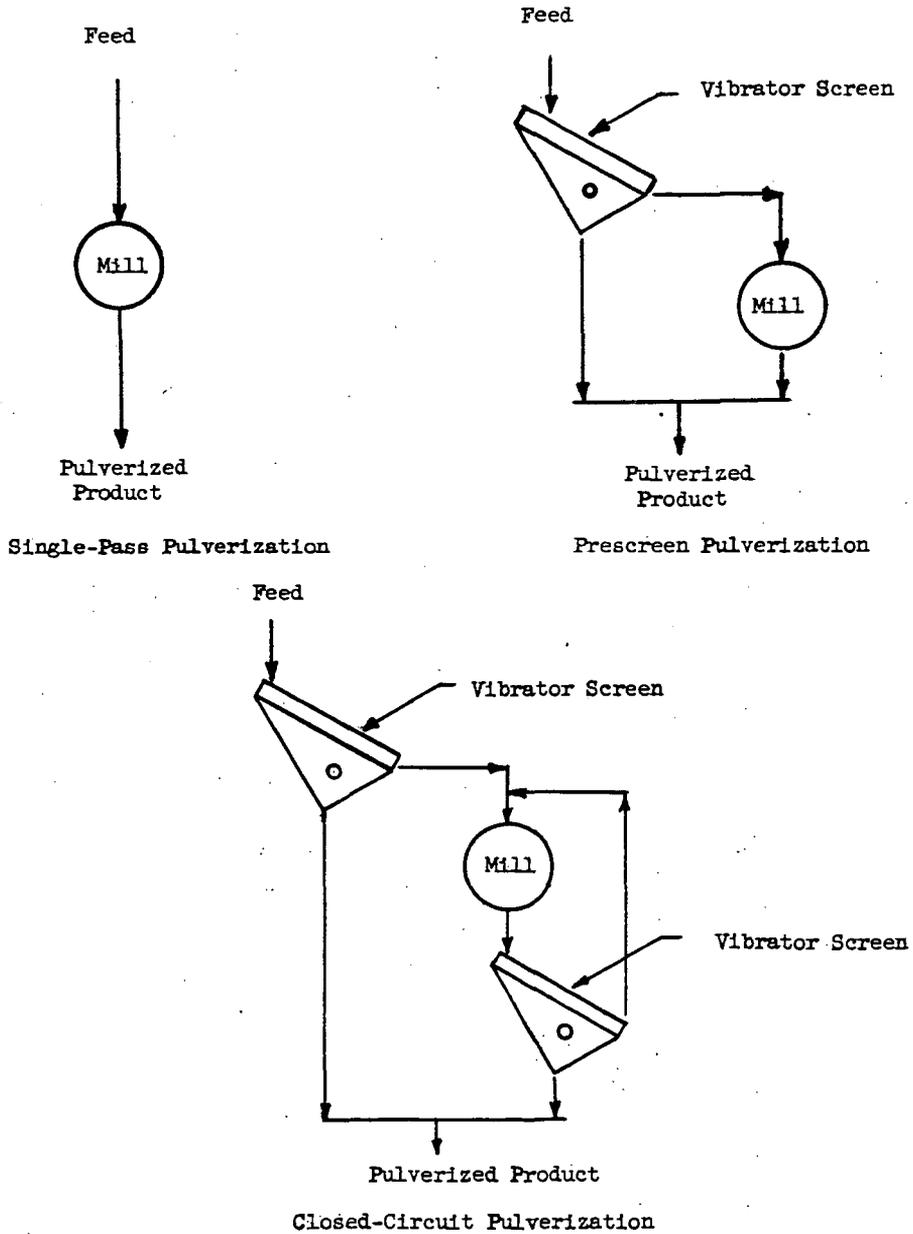
In this review it has been pointed out that in any of the three types of coal-pulverizing systems, mill speed and mill feed rate are the primary variables in controlling pulverization level and bug-dust production. Proper application of these concepts to any of the three systems should lead to practical improvements in pulverization within the limits of the particular system employed.

References

1. A. H. Brisse and H. E. Harris, "Some Basic Principles of Pulverization and Preparation of Coking Coal Blends," Blast Furnace and Steel Plant, 45, p. 301-307 (1956).
2. A. H. Brisse, "Determination of Coke Oven Productivity from Coal Charge Characteristics," Blast Furnace and Steel Plant, 47, p. 376-383 (1959).
3. A. H. Brisse and B. R. Kuchta, "Better Coal Preparation Through Better Crushing," AIME, Blast Furnace, Raw Materials, and Coke Oven Committee, Proceedings, Vol. 18 (1959) pp. 17-21.
4. A. H. Brisse and J. G. Price, "Explorations in Coke Making Research with a Full Scale Coke Oven Model," American Iron and Steel Institute, Chicago Regional Technical Meeting, October, 1959; Blast Furnace and Steel Plant, 47, p. 1285-1290 (1959).
5. J. G. Price and J. D. Clendenin, "Some Practical Aspects of the Degradation of Blast Furnace Coke During Handling," AIME Blast Furnace, Coke Oven and Raw Materials Committee, Proceedings, Vol. 17 (1958) pp. 79-88.
6. C. W. Stahl, "Experiments to Improve Coke Strength," American Iron and Steel Institute Yearbook (1947) pp. 355-378.

7. C. C. Russell, "The Selection of Coals for the Manufacture of Coke," Proceedings American Gas Association (1947) pp. 733-756.
8. C. J. Ramsburg and G. V. McGurl, "Carbonization of Coal-Oil Mixtures," Proceedings American Gas Association (1940) pp. 97, 102-103.
9. W. S. Landers, L. D. Schmidt, and Wm. Seymour, "Control of Bulk Densities in Coke Ovens: Studies on Coal Used at Three By Product-Coke Plants," U. S. Bureau of Mines Report of Investigations 3807 (1945) 22 pp.
10. Pontypridd Test Plant Committee, "A Study of Factors Influencing the Bulk Density of Coal for Carbonization," British Coke Research Assoc. (1957) May, 44 pp.

THREE BASIC TYPES OF COAL-PULVERIZATION SYSTEMS FOR COKE PLANTS



PREPARATION AND COKING RESEARCHES AT BHP CENTRAL RESEARCH LABORATORIES

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INTRODUCTION

The Permian coals of the Newcastle district of N.S.W. yield weak coke, but those mined in the Southern N.S.W. coalfield yield quite reasonable metallurgical cokes. Our problem has been to establish methods whereby stronger cokes can be made from these coals, particularly those from the Newcastle area. Certain aspects of the work concerned with this problem form the subject matter of this paper. A related aspect is dealt with in the paper by J. Gregory to this Symposium.

There are four major phases to the production of metallurgical coke, firstly, the mining and transportation of coals, usually to washeries; secondly, the washing, blending, grinding and delivery of the ground coal to the coke ovens' bunkers; thirdly, charging the ovens, coking and quenching the coke; and, finally, delivery of the coke, perhaps screened and crushed, to the blast furnace. Factors pertinent to the quality of blast furnace coke arise at each stage, and modification of the processing operations at an earlier stage may introduce changes that eventually affect the quality of the blast furnace coke.

Research work can be invalidated by a lack of knowledge of such effects at the production scale or by a failure to reproduce on the smaller research scale, conditions leading to nearly similar materials and processing for the four stages in coke production. In our case, the second (preparation) stage has presented the most immediate possibilities for plant developments. Our research has thus been concerned with this stage to a greater extent than the others.

2. COALS AND PROPERTIES

A summary of the analytical and carbonisation characteristics of the Newcastle coals is given in Table 1. Although Brown, Callcott and Kirov⁽¹⁾ have shown that Seyler's Chart 47B is applicable to Australian coals, the use of Parr's basis to express results is not sound. The commonly used dry-ash-free basis is even less sound, and, so, N.A. Brown and the writer have studied the direct determination of mineral matter and of its water of constitution after modifying the C.S.I.R.O. slow combustion method⁽²⁾. In Table 1, the d.a.f.; Parr and direct dmmf ultimate analyses of "float" and "sinks" fractions of Victoria Tunnel coal are compared. The laboratory carbonisation tests refer to 10% ash washed coal samples. The penetration plastometer is similar to that described by Nadziekiewicz⁽³⁾.

Some or all of the parameters of the laboratory coking tests of the Newcastle coals have been found to depend on the mean particle size and the size distribution, on the proportion of mineral matter and on the extent of oxidation⁽⁴⁾. At present, it seems that the rate of oxidation is one of the few characteristics of these coals that may vary appreciably between the coals⁽⁵⁾.

Results such as those of Table 1 suggest that the bright coals of the Newcastle coalfield are unlikely to yield good blast furnace cokes after conventional preparation. The common coke strength indices of a steelworks blend of these coals, and the better ones made at Port Kembla, are given in Table 2.

The predominant coal minerals⁽⁶⁾ are interlayered montmorillonite-illite, kaolinite, and quartz. The proportions of these minerals depend on the seam and ply; but, for some plies of the seams, they are consistent. The water of constitution of the mineral matter in coal samples varies from 3% to 13%. Much of the mineral matter is finely

disseminated in the coals.

The possibility of "selective" breakage, of one or another form⁽⁷⁾, has been examined. In shatter, no significant selectivity could be detected. Swing hammer mill tests have not so far substantiated any major "selectivity" in breakage; but it seems possible that some differentiation between the minerals and coal may develop. The differentiation was however, never more than slight for feed samples with less than 20% ash and most probably, arose from the differences in density affecting the size classification efficiency of the mill rather than from appreciable differences between the natures of the disintegration patterns for mineral and coal particles or differences in their respective strengths.

3. HAMMER MILL STUDIES

The beneficial effect on coke strength indices of charging finely ground Newcastle coals to coke ovens has long been known. This effect was quantitatively established by subsequent pilot scale and plant scale trials⁽⁸⁾. It was found, however, that plant hammer mills could not attain the required levels of fineness without serious loss in output rates.

The writer has reported⁽⁹⁾ on the early stages of a mathematical analysis of the grinding mechanism of swing hammer mills and experimental studies using high speed cine photography. The breakage in a 5 tph test mill due to the impact of 3¼ lb. hammers at 11000 ft./min. was very mild: for example, a 50% +1 in. product on feeding 100% -2 +1 in. coal. The passage of the hammers past a breaker bar was shown to be equivalent to closed circuit grinding; and the probability of breakage was approximately proportional to the square root of size for sizes smaller than ½ in.; constant at 0.6 for larger sizes. The proportion retained above the breaker bar, for further breakage, varied as the fourth root of particle size. This work suggested that the region above breaker bars (i.e. above the point of narrowest gap between hammer and bar) acts essentially as a means of redirecting particles into this zone. The efficiency of removal of particles which are already sufficiently small, is particularly poor and considerable attention to the design of the recesses above breaker bars may be needed before it is improved.

Work on this aspect and on the design of more suitable screens (grids), after the breaker bars, is in hand. Results to date suggest that a considerable reduction in the proportion of very fine particles in the mill output may be achieved.

Fig. 1 shows a typical path for the discharge of a 1mm particle from a model of a hammer mill fitted with grids. The hammer tip speed was much slower than normal hammer tip speeds. These photographic studies showed that the classification efficiency will remain low so long as the particles must change substantially their direction of flight. This feature also lowers mill capacity; and increases the percentage of fines by undesirable further breakage of particles already sufficiently small.

One alternative to the use of different types of grids is to replace them by adjustable breaker bars and recesses that efficiently redirect particles. The Hazemag mill may be a step in mill development along these lines. It remains to be seen whether the scavenging efficiency of hammers sweeping past bars can be drastically improved.

An alternative to the improvement of hammer mill grid systems is the coupling of a mill with an external classifier.

4. AIR CLASSIFICATION FOR COKING

Screens as external classifiers for hammer mills have been used in Europe to prepare coal for charging coke ovens⁽¹⁰⁾. It appears possible that some additional benefits and more flexible operation may attend the coupling of a suitable air classifier with a hammer mill.

A small commercial air separator, made available by courtesy of International Combustion Australia Ltd., was modified at the B.H.P. Central Research Laboratories, (hereafter CRL). After a number of trials and adjustments, it was found that, at various settings, separations with the efficiencies shown in Fig. 2 could be attained; but

reliable estimates of the potential capacity of such units were not possible with this equipment. A convenient cheap unit (Fig. 3) was made to estimate capacity and confirm the separation efficiency results.

From the work, the following tentative conclusions were drawn:

- (a) Output rates of about 100 tph (and perhaps more) are feasible with 16 ft. dia. units;
- (b) Particular attention must be paid to the evenness of the air distribution around the coal distributor plate;
- (c) Coals containing 2 in. lumps can be handled;
- (d) Total moisture contents up to 12% can be handled without detectable loss in separation efficiency, but the coal should be oiled (about $\frac{1}{4}$ gal./ton) and a special scraper device is probably essential to prevent build up on the wall.

Since this work was based on a 2'6" dia. unit and used feed rates of up to 20 tph, it is far from fully developed as practical plant. However, it does seem one of the possible alternatives to the use of external screens that could be used to control the sizing of system outputs. Such pneumatic controls may prove more stable and consistent than screen and mill or normal mill systems for the preparation of charges to plant coke ovens.

Since this work on an air classifier was completed, an article on the pneumatic preparation of coking coals has been published⁽¹¹⁾. It seems that the system used by Lazovskii et al. is likely to be more easily developed to full plant scale than the external air separator technique.

The 2'6" dia. separators did not break the feed coal to any appreciable extent, but corresponding studies with a small laboratory air separator showed that considerable breakage occurred when feeding $\frac{1}{8}$ in. coal. This feature has prevented the use of this unit in preparing coal for small scale coking tests.

5. BULK DENSITY AND SIZE SEGREGATION

The influences of surface moisture and particle size on the bulk density of particulate materials are well known. In coking practice, the interrelations between these three factors become especially important where, one and perhaps more of the factors strongly affect coke strength. Fig. 4 shows the relationship between bulk density (ASTM Box Test) and fineness (% -18 BSS) for Newcastle coals tested at the B.H.P. steelworks. Total moisture was fairly constant. Fig. 5 shows the corresponding relation between fineness and the Rosin-Rammler distribution coefficient. Stability and hardness indices of cokes from the coals increased by 5 ± 3 and 2 ± 1 units, respectively, for every extra 10% in coal fineness. Another factor may also have entered into the complex of relations: segregation of particles with respect to size in the coke ovens.

Bulk density tests with air dried $\frac{1}{8}$ in. coal have shown evidence suggesting a dependence of bulk density on feed rate and accompanying changes in size segregation. Fig. 6 shows the "herringbone" segregation pattern developed on allowing randomly mixed coal (large) and limestone (fines) to gravitate through various rectangular slots. The bulk density of the discharged material did not depend on the feed rate (i.e. slot width). It has not been possible to generalise these and other observations as regards segregation in coke ovens, but "herringbone" segregation may occur in large ovens.

A direct assessment of the effect of segregation on coke strength was attempted with C.R.L.'s 15 lb. coke oven (see later). The stability and hardness indices of the cokes from segregated and non-segregated charges were almost identical; but the textures of coarse and fine bands were clearly distinct. In view of the marked effect of coal size on these indices, work to elucidate the apparent paradox is being carried out.

6. THE USE OF 15 lb. TESTING OVENS

Pilot (850 lb.) coke ovens are used at the Newcastle and Port Kembla steelworks' research departments, but the associated coal preparation facilities⁽¹²⁾ and coking units are too large for the Central Research Laboratories. Small testing ovens⁽⁸⁾ (Figs. 7, 8) that coke 15 lb. of coal, have therefore been developed. At this scale, many different preparation methods are easily studied and factors that normally interact (e.g. fineness

and bulk density), can be controlled independently of each other. These small ovens, also, provide an easy means of carrying out preliminary surveys prior to confirmation of important observations at the steelworks.

The correlation of the cokes from the small ovens with those from plant and pilot ovens is not yet completed; but appears good (Fig. 9). The reliable prediction of plant coke qualities from those of 15 lb. oven cokes, also, necessitates a similar dependence of coke qualities on various factors.

A series of 15 lb. oven tests to evaluate blending effects and those of certain preparation factors has separated the relative significance of coal fineness and charge density, as well as confirming the usefulness of this small test for complex blending studies.

The preparation of three blends (Table 3) of either 8 or 7 coals is outlined in Fig. 10. The reproducibility of the preparation of the coals' samples is shown, by the ash variance (between samples within coals) of 0.16% (15 deg. freedom), to have been satisfactory. Average ash percentages of washed coals correspond to those of Table 1. The main advantages of the preparation methods were: reduction in amounts of coal washed; the easy prevention of oxidation during storage, and the ease of dewatering coal samples before blending.

The high reproducibility of the characteristics of oven charges is shown by the variances in Table 4. Rotary sample dividers (20 and 100 lb. capacity) were used. Further evidence for the adequateness of the overall reproducibility of the coke tests, is shown in Table 5. It appears from the significance of coking time variations ($\pm \frac{1}{4}$ hr. test to test), that greater control over the rate of heating and the wall temperature-time relationship is desirable.

The separate effects of fineness and charge density are illustrated by Fig. 11. Apart from coking time (possibly a measure of heating rates), other variables were not significant by virtue of either their comparative constancy or not influencing coke qualities.

The effects of the controlled factors (coal fineness and charge density) are similar to those observed during plant and pilot-scale coking tests⁽⁸⁾. They also agree with the trends deduced⁽⁸⁾ from certain American studies. As a rough rule, each additional one percent of coal fineness (% -18 mesh) leads to an increase of about $\frac{1}{4}$ stability units for higher volatile blends of both Australian and American coals.

The very close equivalence of the qualities of the cokes from the four blends confirms the obvious deduction from Table 1: the Newcastle coals are too alike for blend proportions to be very important. This position appears to apply at the steelworks, provided the percentages of ash for all components are reasonably close.

Since the 15 lb. oven yields only about 10 lb. of coke from the high volatile Newcastle coals, some modifications of the ASTM tumbler test have been adopted. The drum is the same as the ASTM drum except its length is 6 ins. The coke charged to the drum is 90% -3" +2" and 10% -2" +1 $\frac{1}{2}$ " and the weight of the coke feed is 3 1/3 Kg. Results show that these changes do not significantly affect the stability and hardness indices for cokes from the Newcastle and Port Kembla steelworks; but, for run-of-wharf coke, the reproducibility is worse than when using the standard test. The reproducibility using cokes from duplicate oven tests appears better than is obtainable with 3 1/3 Kg samples of run-of-wharf coke.

The 15 lb. oven studies, which have involved well over 100 runs in recent months indicate the utility of small ovens. Firstly, they have shown that small scale preparations can be closely controlled and that the greater independence between preparation factors, achievable with small scale work, can lead to results that facilitate the appreciation of results gathered by pilot or plant scale work. Secondly, they have shown that with proper control, 15 lb. ovens can be used for preliminary studies and, in the future, they may enable the restriction of 850 lb. ovens to work immediately prior to

plant trials. The successful use of these ovens depends on, (a), proper control over preparation and charging, and, on (b), a controlled coking programme appropriate to the type of coal e.g. high or low volatile. Our observations in this regard suggest some less specific needs than those of Price et al(13); but there is a large measure of agreement.

ACKNOWLEDGEMENTS

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REFERENCES

1. N.A. Brown, T.G. Callcott, N.Y. Kirov: Paper A6. Preprint. Inst. Fuel Symposium on 'Australian Fuels and Their Utilisation', August 1959.*
2. Coal Research in C.S.I.R.O.; 1959, No. 7, p.13.
3. Nadziekiewicz: Coke and Gas, 1957, May, p.193.
4. T.G. Callcott, P.L. Muir, J.A. Innes: B.H.P. Tech. Bull., 1960, 4, No. 1,
5. P.L. Muir: Paper A8 Aust. Symposium.
6. T. Thompson: B.H.P. Tech. Bull., 1959, 3, No.2, p.20 (also pte. comm.)
7. T.G. Callcott: Paper No.16; Symp. "Sci. in Use of Coal" Inst. Fuel, 1958.
8. T.G. Callcott and R.H. Jones: Paper B10, Aust. Symposium.
9. T.G. Callcott: Paper B5, Aust. Symposium.
10. E. Burstlein: Chaleur et Industrie, Dec. Jan., 1954-55.
11. Lazovskii et al; Koks i Khimia; 1959, No.6, pp.5-8.
12. J.A. Gregory: B.H.P. Tech. Bull. 1959, 3, No. 1, p.23.
13. J.G. Price et al: Proc. Blast Furnace, etc. Ctee., 1959, Vol.17.
14. J.A. Gregory: Paper B11 Aust. Symposium.

* Paper, Preprint. Inst. Fuel. (Aust. Membership). Symposium on 'Australian Fuels and Their Utilisation', August 1959, will be given as "Paper, Aust. Symposium." in refs. 5, 8, 9, 14.

TABLE 1: CHARACTERISTICS OF NEWCASTLE COALS, NORTHERN N.S.W.

COALS SEAMS: Borehole, Victoria Tunnel, Young Wallsend, Dudley.

COLLIERIES: Burwood, John Darling, Stockton Borehole, Lambton.

SIZE GRADING EX BRADFORD BREAKER: $0.74 \leq x < 1.06$ in.; $n = 0.75 \pm 0.05$ (Rosin-Rammler Eq.)

Seam	Percent Ash of Floats at 1.45 S.G.
Borehole; Dudley	$6\frac{1}{2}$ to 10% (d.b.)
Victoria Tunnel	$10\frac{1}{2}$ to 12% (d.b.)
Young Wallsend	13 to 14% (d.b.)

AVERAGE ANALYSES WASHED COALS (7 TO 14% ASH ALL COALS)

BASIS	V.M. %	C %	H %	N %	S % (org)	O + Errors
d.a.f.	39.4	83.5	5.6	2.1	0.45	8.35
Parr dmmf	38.7	84.85	5.55	2.15	0.4	7.05
Direct dmmf	38.8	84.4	5.55	2.2	0.4	7.45

COMPARISON OF BASES FOR FLOATS AND SINKS (Victoria Tunnel Seam)

BASIS	dry basis		VME%	C%	H%	N%	S%	O+Error	δv (Seyler)
	% Ash	% LM							
Floats	daf	11.2	38.9	83.6	5.52	1.96	0.36	8.49	
	Parr dmmf		12.5	58.1	84.9	5.48	1.98	7.31	1.2
	Direct dmmf		12.1	38.3	84.5	5.49	1.98	7.67	0.6
Sinks	daf	51.8	41.8	76.4	5.50	1.84	0.40	15.85	
	Parr dmmf		58.2	35.4	88.1	5.17	2.12	4.21	5.7
	Direct dmmf		56.4	38.4	84.1	5.50	2.03	7.75	0.5

LABORATORY CARBONISATION TESTS (c.10% ASH d.b. ALL COALS)

B.S. SWELLING INDEX GIESELER PLASTOMETER		7½	GRAY KING COKE TYPE AUDIBERT-ARNU DILATOMETER		G4/G5
Initial Move.		380°C	Initial Move.		396°C
Fusion		415°C	Max. Contraction		30%
Log. Max. Fluidity		2.7-3.0	Max. Contraction		438°C
Max. Fluidity		435°C	Ult. Dilatation		40%
Resolidification		465°C	Ult. Dilatation		465°C
<u>PENETRATION PLASTOMETER</u>		(0.6 Kg/cm ²)			
Layer Thickness	25 mm	Top Temps.	330°C		
Shrinkage	22 mm	Bottom Temps.	440°C		
<u>HARDGROVE INDEX</u>		55			

TABLE 2: COKE STRENGTH

Coke	Percentage Index		+½in. Hardness
	+1½in. Shatter	+1in. Stability	
Newcastle (Northern N.S.W. Coals)	73	18	63
Port Kembla (Southern N.S.W. Coals)	85	45	60

TABLE 3: BLEND PROPORTIONS (DRY WEIGHT %)

COAL	BLEND		
	No. 1	No. 2	No. 3
A	6.1%	NIL	NIL
B	10.1%	13.0%	12.2%
C	9.1%	19.7%	8.5%
D	16.1%	13.5%	13.7%
E	15.85%	14.1%	13.9%
F	12.0%	10.7%	12.1%
G	20.6%	20.8%	31.0%
H	10.15%	8.3%	8.8%

TABLE 4: REPRODUCIBILITY OF OVEN CHARGES

	Ash % (d.b.)	Moisture % (total)	% -18 BSS	Charge Density (lbs./c.ft.)
Variance	.04	.06	2	1/4
Deg. Freedom	60	49	20	30

TABLE 5: ANALYSIS VARIANCE OF FACTORIAL DESIGN FOR TESTS OF TWO BLENDS

Factors: a, 43% and 73% -18 BSS; b, 37, 43, 49 lbs./cft; c oven No.1 & 2, d, Blend 2 & 3

Effect	Stability Index Mean Square	Hardness Index Mean Square
Grand Mean	19,136.55	2470.5
Fineness (A)	444.6 (Sig)	313.2 Sig
Ch. Density (B ₁)	0.04	194.6 Sig
Ch. Density (B ₂)	8.7	2.0
Ovens (C)	7.2	2.87
Blend (D)	73.8 (Sig)	0.01
Coking Time**	17.9 (Sig)	N.S.
Res M/s	2.568; 10 d.f.	1.91
Total	19,755.17	3018.41

NOTE: Mean square of each interaction (e.g. AB₁D) was estimated separately and unless specified was not significant at 10% level.

** If coking time significant, at 5% level; reduced res. m/s is given and mean squares of other effect carried to average time.

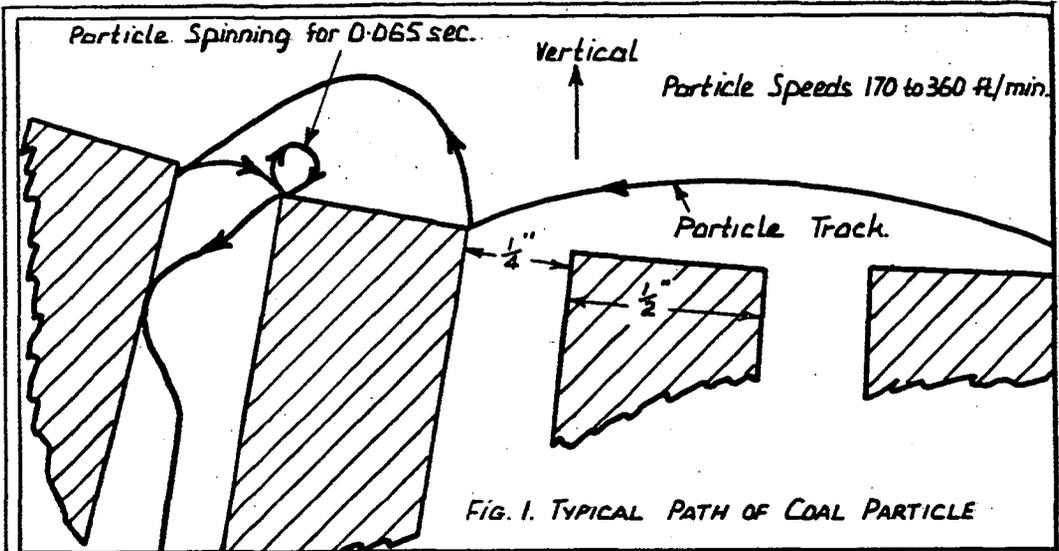


FIG. 1. TYPICAL PATH OF COAL PARTICLE

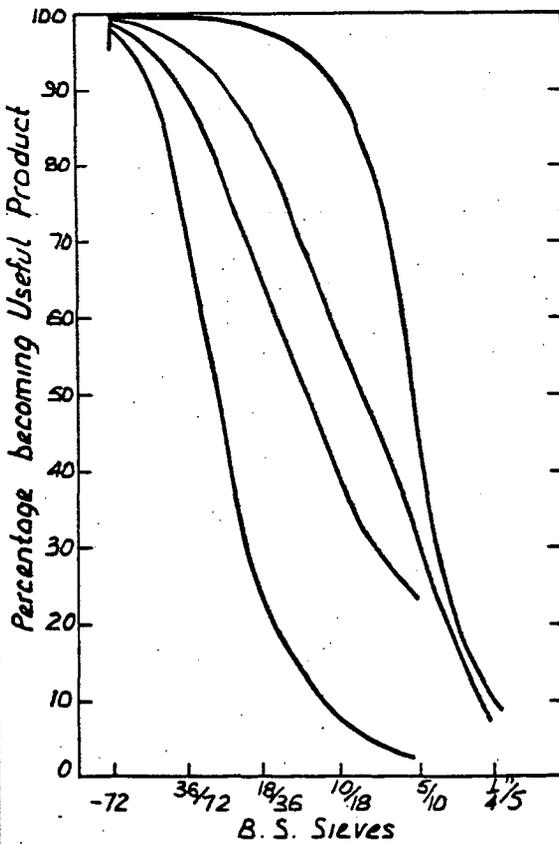


Fig. 2. CLASSIFIER EFFICIENCIES.

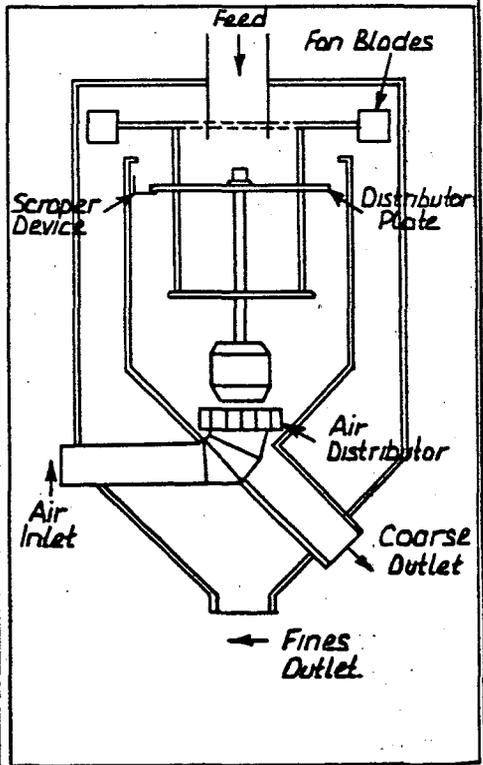
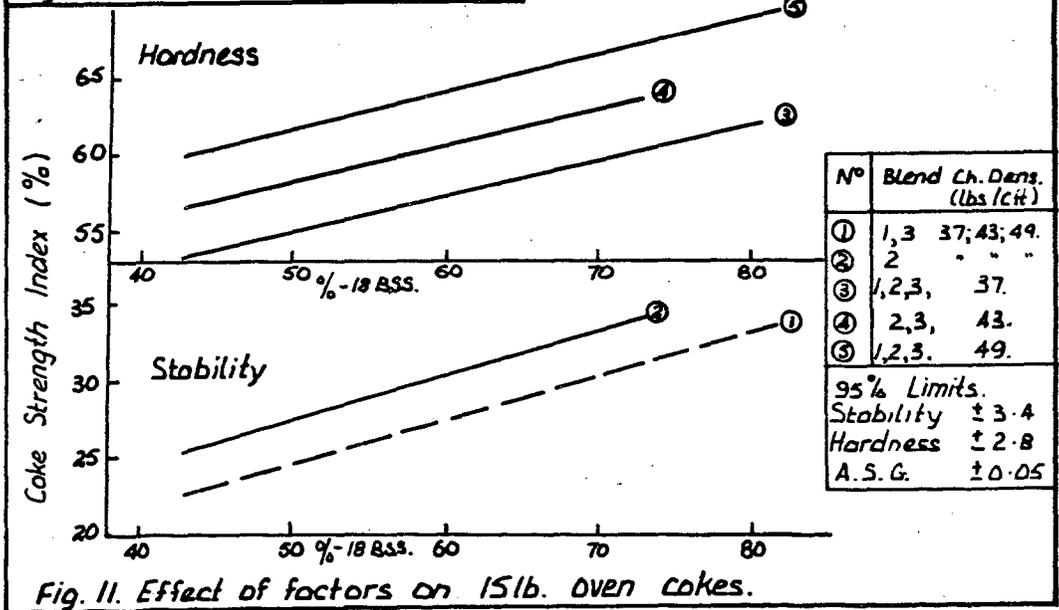
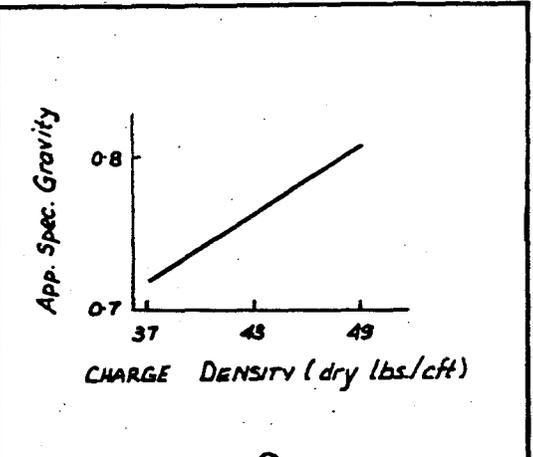
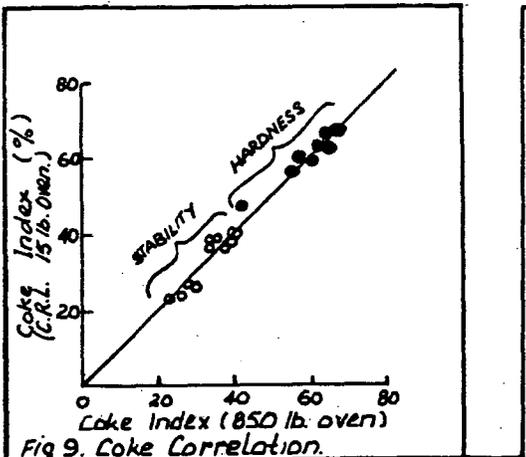
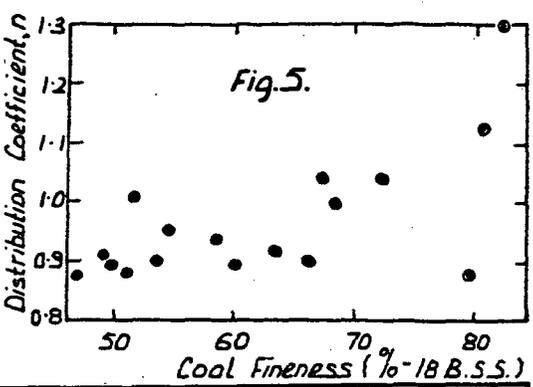
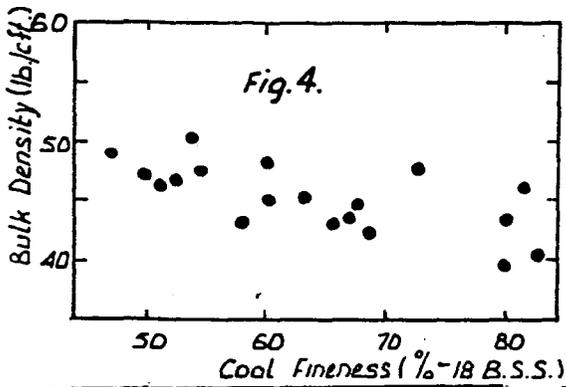


FIG. 3. C.R.L. AIR SEPARATOR



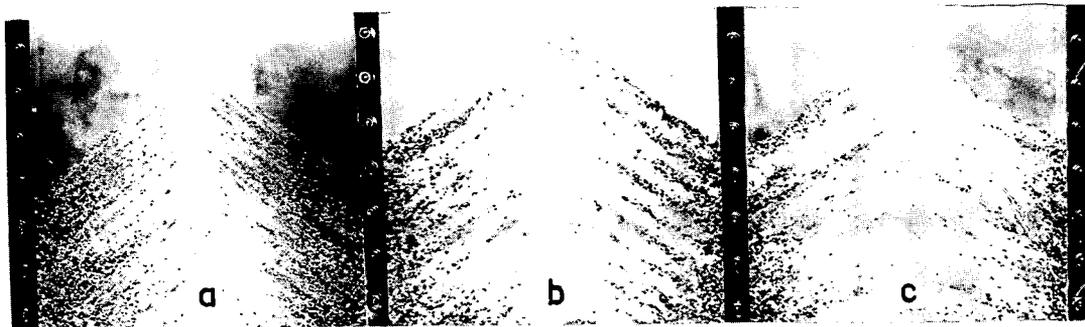


Fig. 6. Segregation Patterns. Discharge Apertures $a = \frac{1}{4}$ ", $b = \frac{1}{2}$ ", $c = 1$ ".



Fig. 7. Coke From 15lb. Oven.

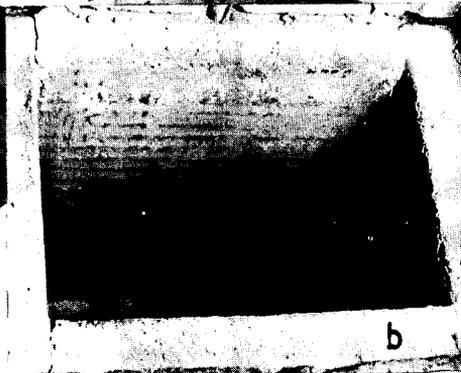
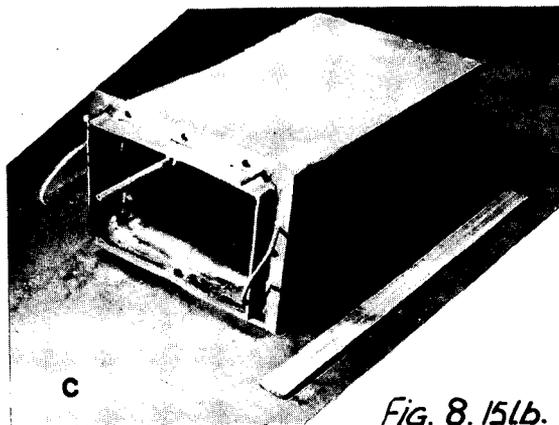
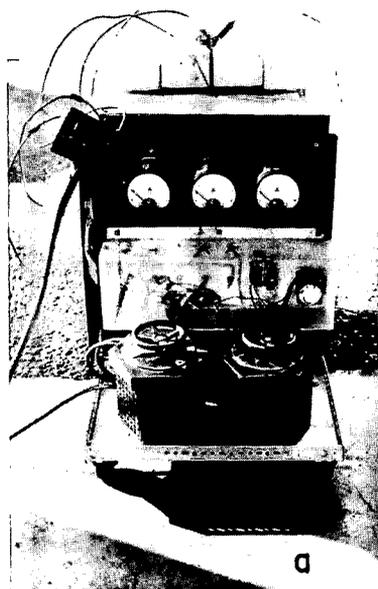
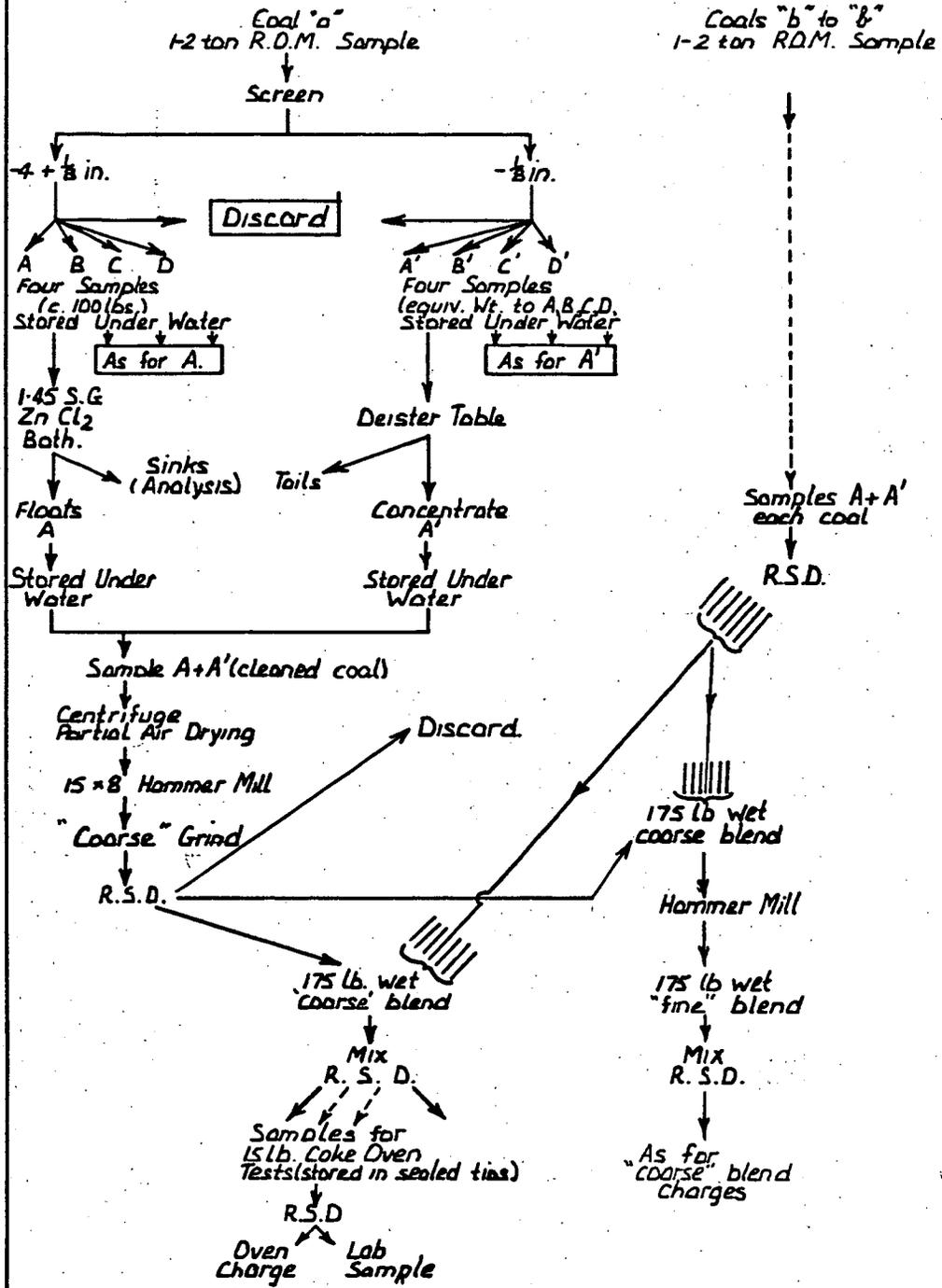


Fig. 8. 15lb. Test Coke Oven
a. Assembled Oven.
b. Oven Chamber
c. 25/20 Stainless Steel Retort.

FIG10. PROCEDURE: PREPARATION BLEND OF 8 COALS.



PLANNING OF CENTRAL WASHERIES IN INDIA
FOR PREPARATION OF COKING COALS*

by

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ABSTRACT

This paper is expected to cover washing problems peculiar to the coals of India which are commonly high in mineral matter content. This characteristic has required the application of two-product** washing practice to achieve production of acceptable metallurgical coals. The middlings product is used for generation of steam and electricity. Central washeries require most careful design*** and planning of all the interrelated operations of mining, and transport of the raw coal, washing, disposal of refuse, and shipping of the product to maintain satisfactory operation.

* Manuscript not received in time for preprinting.

** A. Farquhar, "Beneficiation of Jharia and West Bokaro Coals by Washing" Progress of the Mineral Industry of India (1906-1955) Golden Jubilee Commemoration Volume, The Mining, Geological and Metallurgical Institute of India, Vol. 21, Chowringhee, Calcutta 13, pp. 206-250.

*** G. G. Sarkar, S. Sarka and A. Lahiri: Journal of the Institute of Fuel, Vol. 33, No. 230 (1960) March pp. 145-150.

PLASTIC PROPERTIES OF SUNNYSIDE COAL.
CLEANING PLANT PRODUCTS, OXIDATION AND STORAGE
BLENDS WITH OTHER COALS AND WITH OTHER MATERIALS

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Coal from the Kaiser Steel Corporation's Sunnyside, Utah, Mines has been the principal source of coal for making blast furnace coke at Fontana since the start of operations in 1943. Requirements have increased from two batteries of coke ovens supplying one blast furnace to seven batteries of ovens making coke for four blast furnaces. There have been many changes in mining methods, in coal preparation, and in the use of the Sunnyside coal, but this coal still remains the major source of coking coal for the Fontana plant. Experience in use during the years, as well as laboratory investigations, has provided considerable information on the properties of the Sunnyside coal and its behavior during carbonization, both alone and in blends with other materials. These investigations are continuing to provide better coke, both with respect to chemical properties such as ash and sulfur content, as well as for the physical properties. Better coke gives increased iron production, lower coke rate, and the ultimate object, reduced cost of hot metal.

Early operations at Fontana showed that satisfactory blast furnace coke could not be produced from straight Sunnyside coal.⁽¹⁾ It was necessary to obtain other better coking coals or other materials for improving the size and strength of the coke and to reduce breeze. Changes in coal supply have included purchase of extensive properties in the Raton field in northern New Mexico and use of Koehler mine coal from there. Sources of supply have been developed of purchased medium and low volatile coals for blending with the Sunnyside coal to improve the size and strength of the coke.

It is common practice to blend low volatile coal with high volatile coal for increasing the size and strength of the coke.⁽²⁾ However, there is no production of low or medium volatile coal nearer to Fontana than the Crested Butte field in Colorado or the interior province coals of the Arkansas-Oklahoma region. Coals from these areas must be shipped by rail; thus the freight cost becomes a very important factor in the economy of operations. Details of the coal handling practice at Fontana have been described recently.⁽³⁾

MINING AND COAL PREPARATION

The Sunnyside coal is produced from three separate adjoining mines. Raw coal from all three mines goes to mixing bins to provide a feed as uniform as possible for the coal cleaning plant. The combined coal is crushed to a top size of six inches and the 6 x 0 coal passes through jigs for cleaning. The treatment of the float coal from these jigs for dewatering results in several sizes of products. The 28 mesh x 0 fines from the screens are further treated by flotation and the cleaned coal product is recombined with the coarse clean coal for shipment to Fontana. Details of the flotation cleaning plant have been published.⁽⁴⁾

All of the cleaned coal is shipped immediately and in normal operation no coal is stocked at the mine. Because of the practice of recombining all size fractions of cleaned coal as well as no stocking at the mine for dewatering or drying, the coal arrives at Fontana with 6 to 7% total moisture, the flotation product itself containing 20 to 25% moisture. At Fontana all of the coal received goes to stock and stock coal, 45 - 60 days old, is used in the blend. Fears that Sunnyside coal could not be held in stock even for this length of time without serious decrease in the

already poor coking properties has not proven true in practice.

LABORATORY TESTS OF COKING PROPERTIES

Full-scale testing of possible blends is both difficult and expensive, so that laboratory tests to permit selection of the more suitable coals for blending is highly desirable. A variety of laboratory methods have been described in the literature⁽⁵⁾ for assessing the coking properties of coals, ranging from tests on usual laboratory samples as small as one gram to various pilot scale test ovens using 500 lbs. or more per test.⁽⁶⁾⁽⁷⁾ In general, laboratory methods, using rapid rates of heating, are unsatisfactory for application to poorly coking western coals. Often such tests yield cokes, whereas testing at usual rates of carbonization does not yield a coherent cellular coke residue. The Gieseler plastometer test, generally following the proposed ASTM method,⁽⁸⁾ has been found most useful for western coals. An illustration of Gieseler plastometer test results is given in Chart I for three different types or ranks of coal. This illustration shows differences in maximum fluidities attained during the test as well as differences in the temperature ranges during which plastic properties were indicated.

Besides the Gieseler plastometer test, both the free swelling and the agglutination tests⁽⁹⁾ have been used at times. Results for these tests, compared to values for the maximum fluidity shown in the Gieseler test, are given in Table I and shown graphically in Charts II-A and II-B. The tests for these comparisons at various levels of plasticity were done on 300 g samples of -35 mesh coal. The original sample was tested in the plastometer and portions taken for further grinding to -60 mesh and -200 mesh for the free swelling and the agglutinating test. The remaining gross portion of the sample was placed in a gravity convection drying oven at 105°C. After a selected time interval the sample was removed, cooled, and a portion taken for plastometer testing and other portions for grinding and testing by the other methods. This was repeated until the extent of oxidation was such that no coherent residue was obtained in the Gieseler plastometer test.

The results showed different relationships for different ranks of coal. For purposes of comparison it was found that, if the maximum fluidity in the Gieseler test was less than 2 dial divisions per minute, the residue remaining after the test was only a sintered button and did not exhibit the cellular structure associated with coke. Thus, it was concluded that neither the agglutinating test nor the free swelling test are satisfactory for assessing the coking properties of coals, but could be used possibly to indicate major changes in the coking properties of a coal if the general properties were known from Gieseler plastometer and other testing.

PLASTIC PROPERTIES OF VARIOUS SIZES OF SUNNYSIDE COAL

Plastometer tests were run on each of the sizes of coal obtained at the cleaning plant. The samples were taken at Sunnyside and were 10 to 15 days old when tested at Fontana. The data obtained, Table II, showed the expected increase in moisture with decrease in particle size. Other trends included decrease in volatile matter and increase in ash. The Gieseler plastometer results showed decidedly lower plastic properties for the finer sizes.

Petrographic analyses of the Sunnyside coal had indicated that the coal was over 90% clarain or Type III vitrinite. There were no appreciable percentages of harder or softer constituents such as durain or fusain that might be expected to segregate in the larger or finer sizes and to cause different coking properties by segregation. Therefore, it was indicated that the finer sizes of coal produced at the mine had already undergone considerable weathering between mining and delivery and testing at Fontana.

OXIDATION TESTS

Results at this time were somewhat confusing. Although preliminary studies had indicated that the Sunnyside coal was susceptible to rapid weathering in stock, actual plant practice had shown little change in the coal in 60 days or often a longer period of time. Gieseler tests of stock coal had shown considerable variation in maximum fluidity, Table III, but no apparent trends; average values were not particularly lower than averages for fresh incoming coal. Plastometer testing also was giving confusing results. Duplicate testing, both to check individual samples and to check the results obtained by different operators, did not show good agreement. Further, tests of a series of samples such as blends of Sunnyside with other coals did not show consistent results. Normal fresh Sunnyside coal had been found to give maximum fluidities of the order of 40 to 80 divisions per minute. In the test data shown in Table I the original sample had a maximum fluidity value of only 19 and two hours at 105°C had decreased this value to only 5.7 divisions per minute, indicating rapid oxidation.

It was decided to make a careful examination of the change in plastic properties with time for -35 mesh Sunnyside coal at room temperature. A sample of about 150 grams of -35 mesh Sunnyside coal was prepared as rapidly as possible and then tested immediately. The sample was then set aside in a rubber stoppered glass bottle and tested weekly. The results of this test work are shown in Chart III. As shown in this chart, only one week in this condition had decreased the maximum fluidity from 60 to only 21 divisions per minute. Subsequent testing showed further considerable decline in maximum fluidity but at a decreasing rate. In view of these results a second sample was prepared and tested daily. The results showed a decrease in maximum fluidity from 60 to 33 with only 24 hours standing at room temperature, and further decrease to about 20 after one week. These results explained the lack of agreement in duplicate testing and may explain in part the considerable variation shown in the maximum fluidity of plant control samples. In view of these results a standard time program of sample preparation and of testing was formed.

A variety of methods have been described in the literature for preserving samples of coal for future study. These include storage under an inert atmosphere such as natural gas, filling the air space in the container with water, and freezing the samples. Except for the use of an inert atmosphere, these methods often would require further treatment of the sample that may produce changes as great as would have occurred during storage under air. Also, as shown in Chart II, storage under natural gas served only partially to deter the oxidation and the decrease in the plastic properties.

Because six samples is about the maximum number that can be tested by one operator in one plastometer in an 8-hour work day, and because of the various difficulties of preserving samples, a group of tests was run to study the change in plastic properties with respect to particle size of the coal being preserved. The results are shown in Chart IV. The data obtained indicated that 1/4" was the minimum particle size for a sample that was to be held even for the few days of a testing program.

It may be noted at this point that some of these same difficulties were experienced in coking tests with a 500-lb. test oven. It is customary to obtain a relatively large sample of coal for use in a series of tests in order to eliminate variations that might occur in separate samples. It was found that Sunnyside coal, ground in the laboratory hammermill to approximately 75% -1/8" and preserved in covered steel drums, showed decided changes in coking properties within two weeks storage.

PLASTIC PROPERTIES OF BLENDS OF SUNNYSIDE WITH LOW VOLATILE COALS

Only after the rapid change in plastic properties of -35 mesh Sunnyside coal at room temperature was understood was it possible to obtain satisfactory results for plastic

properties of blends of Sunnyside coal with other materials. As shown previously in Chart I, the plastic temperature range for Sunnyside coal extends from about 335° to 460°C. Many true low volatile coals, on the other hand, show plastic properties in the Gieseler test in the range 440 to 505°C. Only a small temperature interval of perhaps 20° is common to these two types of coal.

The change in maximum fluidity with change in composition of blends of Sunnyside with two different low volatile coals is shown in Chart V. The maximum fluidity shown for the Sunnyside coal and low volatile coal "A" was approximately the same at 50 to 60 divisions per minute. However, adding this low volatile coal to Sunnyside coal produced a rapid decrease in the maximum fluidity with increase in the percent of the low volatile coal used to a minimum of approximately 2½ divisions per minute at a composition of about 75% Sunnyside and 25% low volatile coal. The second series of tests was run using low volatile coal "B" of much lower maximum fluidity. The same type of curve resulted except that the change in maximum fluidity with increase in percent coal "B" was greater than when the more fluid low volatile coal "A" was used.

A minimum value for the fluidity of a blend of coking coals cannot be given because the quality of the coke is influenced by many other factors besides fluidity. However, plant practice had shown that at times during the year, particularly in the fall, the coke quality decreased. This decrease in quality with increase in breeze was found to be related to the decrease in the maximum fluidity shown by the mixed coal being carbonized. Attempts to improve the coke quality by increasing the percentage of the low volatile coal were not very successful and the reason for this is shown by the course of the curves given in Chart V. Increase in the percent low volatile coal decreases the fluidity of the mixed coal.

PLASTIC PROPERTIES OF BLENDS OF SUNNYSIDE WITH MEDIUM VOLATILE COALS

Medium volatile coals are less desirable from the economic standpoint for blending with Sunnyside coal. Higher percentages of medium volatile coal are required to obtain a given increase in the size and strength of the resulting coke. The carbon return in carbonization is lower than obtained from low volatile coals. Nevertheless, the generally excellent coking properties of medium volatile coals have been found to be desirable for use at Fontana for obtaining better fused cokes and reduction in breeze.

The results of Gieseler plastometer tests of blends of Sunnyside coal with two different medium volatile coals are shown in Chart VI. It may be seen that the change of maximum fluidity with change in composition is not linear. For practical purposes only the parts of the curves from 0 to 20% medium volatile coal additions have been of practical interest. Additions of medium volatile coal in this range cause little increase in the maximum fluidity of blends with Sunnyside coal. However, in comparison with the use of low volatile coal, it is significant that the maximum fluidity actually increased or remained the same and did not show the very marked decrease of Sunnyside-low volatile coal blends.

PLASTICITY RELATIONSHIPS USING KOEHLER COAL

The Koehler coal from the Raton, New Mexico, field had been used at various times even before purchase of the property by the Kaiser Steel Corporation in 1957. The Koehler coal shows considerably better plastic properties than the Sunnyside coal although the proximate analysis indicates approximately the same rank of coal, Table IV. Also, coke produced from straight Koehler coal is appreciably larger and stronger than is obtained from Sunnyside coal. The Gieseler plastometer test shows approximately the same temperature range of occurrence of plastic properties. Blends of Sunnyside and Koehler coal showed a linear change in maximum fluidity with change in composition, as illustrated in Chart VII. Blends of Koehler coal with low

volatile or with medium volatile coals showed the same general changes in maximum fluidity with change in composition as described for the blends using Sunnyside coal. Thus, in general, Koehler coal can be substituted for Sunnyside coal.

PLASTIC PROPERTIES OF BLENDS OF LOW AND MEDIUM VOLATILE COALS

Tests were made of blends of medium volatile and low volatile coals for basic information of blending. The results are shown in Chart VIII. As would be expected, due to the great variety of both low and medium volatile coals, a great number of combinations are possible. The results shown in Chart VIII represent only two series of possibilities. These results indicated that addition of medium volatile coal to low volatile coal produced appreciable increases in the maximum fluidity when only 20% medium volatile coal was present. This result might be expected from the greater coincidence of the temperature ranges of plasticity compared to blends of Sunnyside with either low or medium volatile coal. Above 20% addition of medium volatile coal very great increases in maximum fluidity were shown although it is probable that these increases are due to a considerable extent to the swelling that occurs during the testing of such blends.

THREE-COMPONENT BLENDS OF SUNNYSIDE, LOW, AND MEDIUM VOLATILE COALS

Plant practice had shown that blends of Sunnyside and low volatile coals were satisfactory only part of the time; at other times the fusion of the blend became poor resulting in excess breeze production and decrease in the tumbler test hardness factor. Use of only medium volatile coal with Sunnyside is not desirable because greater percentages are required for a given level of size and strength. These two considerations resulted in the practice of using both low and medium volatile coals for blending with Sunnyside coal. The relative percentages of low and medium volatile coal are changed, depending upon the economic considerations and the coke requirements at a given time.

Actually, three-component blends of Sunnyside, low, and medium volatile coals result in a variety of curves between the individual two-component series of Sunnyside-medium volatile and Sunnyside-low volatile. Two relationships are shown in Chart IX. When equal quantities of the low and medium volatile coals used were blended with Sunnyside coal, the change in maximum fluidity with change in percent blending coal added showed a decline in maximum fluidity but at a much slower rate than shown by using low volatile coal alone. When the relative percentages were changed to 1/3 low volatile and 2/3 medium volatile coal, the maximum fluidity shown by the blend with Sunnyside coal showed essentially no change in maximum fluidity over the practical range of addition of the blending coal.

OTHER COALS AND MATERIALS FOR INCREASING THE FLUIDITY OF BLENDS

Except for a relatively few areas in Colorado there are no known operating mines in southwestern United States producing high fluidity coking coals. The use of such coals, of course, would be essentially substitution of the high fluidity, probably high volatile, coal for Sunnyside rather than replacement of the expensive medium and low volatile coals being used. Exploration in the Kaiser Steel Corporation's Raton properties has shown extensive deposits of both high volatile coking coal of excellent plastic properties as well as deposits of medium volatile coals also of excellent plastic properties. Use of these sources awaits future development.

Besides other coals, some natural asphalts or bituminous materials are available western raw materials. A number of these materials have been tested. Although such materials become highly fluid during carbonization, not all of them show the same ability to improve the coke quality. It is probable that the differences in effects on plastic properties are related to differences in the plastic ranges of these bituminous materials together with variations in the rate and manner of decomposition

and devolatilization.

One such natural bituminous material that has been used in emergency on the plant scale basis is Gilsonite⁽¹⁰⁾ obtained from northeastern Utah. It was found in plant practice during periods when the coals on hand did not possess sufficient fluidity, that 3½ to 5% Gilsonite could be blended with the coking mix successfully to reduce the plant coke breeze yield. Chart X shows the increase in the maximum fluidity obtained by blending Gilsonite either with Sunnyside coal or with low volatile coal, and comparative results are indicated for the benefits in increasing fluidity obtained using medium volatile coal. It is evident that small percentages of Gilsonite cause decided increases in the maximum fluidity shown by coals in the Gieseler plastometer test.

Chart XI illustrates the application of Gilsonite to a plant coal mix. Addition of 17½% low volatile coal to Sunnyside decreased the maximum fluidity from 42 divisions per minute for straight Sunnyside down to only 20 divisions per minute. Additions of Gilsonite to this 82½% - 17½% blend caused rapid increase in maximum fluidity with increase in percent Gilsonite. Only 4% Gilsonite was required to raise the fluidity of the coal blend up to the value shown by the Sunnyside coal itself. Further small increases in Gilsonite brought the maximum fluidity to over 100 divisions per minute. Carbon return is very low for Gilsonite and the delivered cost is high so that Gilsonite has not been used on a continuing basis.

USE OF INERT MATERIALS FOR IMPROVING THE QUALITY OF COKE OBTAINED FROM SUNNYSIDE COAL

There has always been active interest at Fontana in finding substitutes for the expensive low and medium volatile coals that have been required to improve the coke quality for satisfactory blast furnace performance. It has also been recognized that the possibilities for success were small because Sunnyside coal, coked alone, does not yield completely fused coke. Pebbly seam and, at times, considerable portions of poorly fused agglomerate rather than true coke are found. Thus, this coal could hardly be expected to absorb inert or non-coking carbonaceous materials without further considerable increase in breeze. Even with excellent coking coals such inert materials have been found to give variable results depending on source and composition. For example, some varieties of anthracite have been used successfully for incorporation into coking blends for increasing coke size and foundry coke production at fast coking rates⁽¹¹⁾, but only certain anthracites have been found suitable for this application. At Fontana many varieties of char, anthracite, petroleum coke, and other inert materials have been tested, generally with poor results.

An example of the effects on the plastic properties is the use of petroleum coke as a blending material for increasing the coke size and strength. Chart XII shows the relationships between maximum fluidity of the blend and the percent petroleum coke. As might be expected, increase in percent petroleum coke caused decrease in the maximum fluidity of the blend. However, it is interesting to note that the rate of decrease of the maximum fluidity is less than shown by equivalent additions of low volatile coal. Test work is under way at present to examine the plastic properties of blends of Sunnyside and Koehler coals with inert materials, but using Gilsonite and other natural high fluidity materials for increasing the plastic characteristics of the blends.

CONCLUSION

A knowledge of the plastic properties of the Sunnyside coal has been found useful in explaining experiences in the use of the coal both alone and in blends with other materials. This information has also been beneficial in the selection of other coals for blending and in the determination of suitable blends. However, it is recognized that plasticity is only one factor in the selection and use of coals for carbonization. The Gieseler plastometer test has been found most useful for the

determination of the plastic properties of coals and blends of coals and for the detection of changes in the plastic properties. The understanding of the susceptibility of the Sunnyside coal to rapid changes in plastic properties at room temperature, for fine particle sizes, has been very important for obtaining satisfactory results for duplicate tests and for series of samples. Such rapid change in plastic properties at room temperature may be of importance for other laboratories to consider where difficulties have been experienced in obtaining duplicate results or consistent values for plastometer testing. These effects may become increasingly important in the estimation and use of other higher rank coals because of the increased mechanization of mining and finer coal production, cleaning of finer sizes, the use of heat drying to reduce the moisture content of coking coals before shipment, and the stocking of finer coking coals.

ACKNOWLEDGMENT

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BIBLIOGRAPHY

- (1) Thompson, J.H., Blast Furnace and Steel Plant 34 225 (1946).
- (2) Russell, C.C., and Perch, M., A.I.M.E. Tech. Publ. No. 1520 (1942).
- (3) Schulze, W.G., and Soth, G.C., A.I.M.E. Blast Furnace, Coke Oven, and Raw Materials Conference, April 4-6 (1960).
- (4) Denver Equipment Co., Deco Trefoil 21 No. 3 11 May-June 1957.
- (5) Brewer, R.E., U.S. Bur. Mines Bulletin 445 (1942).
- (6) Harris, H.E., Glowacki, W.L., and Mitchel, J., see ref. 3.
- (7) Lee, G.W., see ref. 3.
- (8) ASTM Standards on Coal and Coke, Appendix III p 129, 1954.
- (9) *ibid.* Standard D 720-57; *ibid.* Sept. 1948, p 151.
- (10) Bond, R.K., Chem. Eng. 64 249 (1957).
- (11) Roberts, I.M., A.I.M.E. Tech. Pub. No. 1612 (1942).

TABLE I

COMPARISON OF GIESELER PLASTOMETER MAXIMUM,
FREE SWELLING INDEX, AND AGGLUTINATION TEST INDEX

SUNNYSIDE COAL: V.M. 40%, ASH 7%, SULFUR 1.2%

<u>SAMPLE</u>	<u>P.MAX.</u>	<u>FSI</u>	<u>A.I.</u>
Original	19	3½	9.1
2 hours at 105°C	5.7	-	-
4 " " "	4.5	3½	6.9
8 " " "	2.9	3½	6.7
16 " " "	1.3	2½	5.5
24 " " "	1.0	2	5.3
36 " " "	.5	1½	3.9
48 " " "	No fusion	1	2.7

LOW VOLATILE COAL: V.M. 18%, ASH 8%, SULFUR .9%

<u>SAMPLE</u>	<u>P.MAX.</u>	<u>FSI</u>	<u>A.I.</u>
Original	13	9½	19.1
4 hours at 105°C	2.8	9	18.5
8 " " "	2.2	9	18.1
16 " " "	1.3	8	15.3
24 " " "	1.2	8	14.3
36 " " "	.8	5½	7.8
48 " " "	.6	4½	4.8
96 " " "	No fusion	3	2.5

TABLE II

ANALYSES AND PLASTIC PROPERTIES OF VARIOUS SIZES OF
SUNNYSIDE COAL FROM THE CLEANING PLANT

SIZE	ANALYSIS (DRY BASIS)					GIESELER PLASTOMETER TEST				
	H ₂ O %	V.M. %	F.C. %	ASH %	SUL. %	F.M. °C	.1 D/M °C	MAX. °C	SOLID. °C	MAX. D/M
6" x 1-5/8"	3.4	42.1	52.4	5.5	1.12	339	370	429	459	55
1-5/8" x 1/2"	4.8	40.3	52.9	6.8	1.18	339	367	429	459	46
1/2" x 3/16"	7.9	40.1	52.9	7.0	1.11	347	369	428	459	19
3/16"x 28 mesh	8.4	39.5	52.9	7.6	1.14	344	372	426	459	14
28 mesh x 0	22.2	39.2	52.6	8.2	1.27	366	387	427	455	8

TABLE III

SUNNYSIDE COAL RECOVERED FROM STOCKPILE
COAL APPROXIMATELY 60 DAYS IN STOCK

SAMPLE NO.	SIZE CONSIST		P. MAX.	
	+28 MESH	-28 MESH	+28 MESH	-28 MESH
1	88	12	38	9
2	84	16	45	9
3	84	16	25	4.8
4	74	26	35	3.3
5	78	22	37	4.9
6	82	18	65	9
7	84	16	35	5.0
8	85	15	45	11
9	82	18	50	7
10	88	12	57	10
11	83	17	32	11
12	81	19	40	10

TABLE IV

PROXIMATE ANALYSES, SULFUR, AND GIESELER TEST DATA

CONSTITUENT	ANALYSIS (DRY BASIS)				GIESELER PLASTOMETER TEST				
	V.M. %	F.C. %	ASH %	SUL. %	F.M. °C	.1 D/M °C	MAX. °C	SOLID. °C	MAX. D/M
COALS:									
Sunnyside	40.4	52.9	6.7	1.21	333	368	426	458	60
Koehler	37.7	49.8	12.5	.71	330	365	427	462	200
Med.Vol.(Okla.)	26.1	66.9	7.0	1.07	377	383	453	504	2700
Med.Vol.(W.Va.)	23.9	69.9	6.2	.58	384	391	467	504	1900
Med.Vol.(Colo.)	23.4	69.1	7.5	.68	386	395	462	507	2000
Low Vol.(Okla.)	19.6	72.3	8.1	.73	425	430	474	511	160
Low Vol.(W.Va.)	18.4	75.6	6.0	.72	425	427	476	510	65
Low Vol.(Ark.)	17.7	74.4	7.9	.88	442	446	482	509	22
Low Vol.(Ark.)	17.3	74.0	8.7	.95	442	446	487	507	7
<u>Petroleum Coke</u>	12.2	87.6	.2	1.15	No Fusion				
<u>Gilsonite</u>	77.7	21.9	.4	.19	Too Fluid To Test				

CHART II-A
COMPARISON OF VARIOUS COKING INDICES
CHANGE OF INDEX WITH OXIDATION-SUNNYSIDE COAL

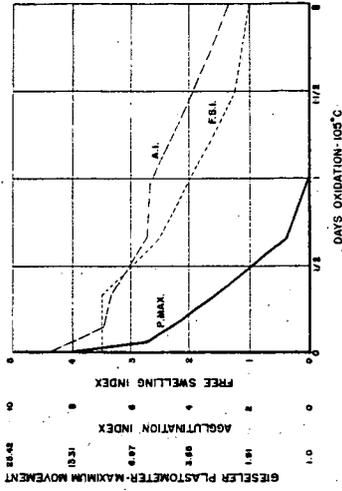


CHART II-B
COMPARISON OF VARIOUS COKING INDICES
CHANGE OF INDEX WITH OXIDATION-LOW VOLATILE COAL

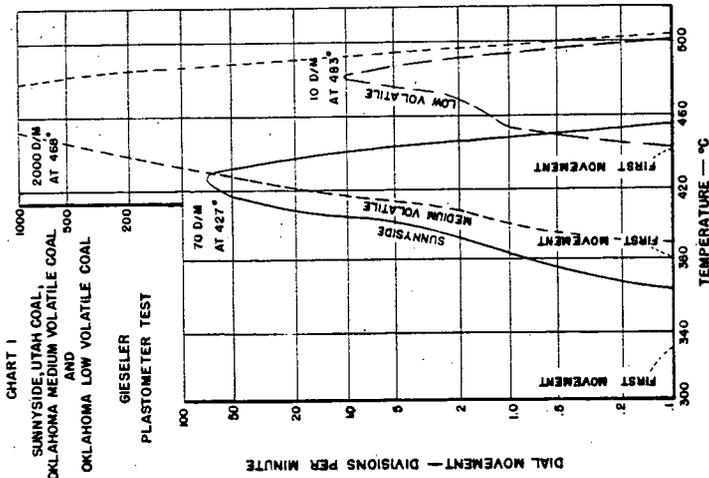
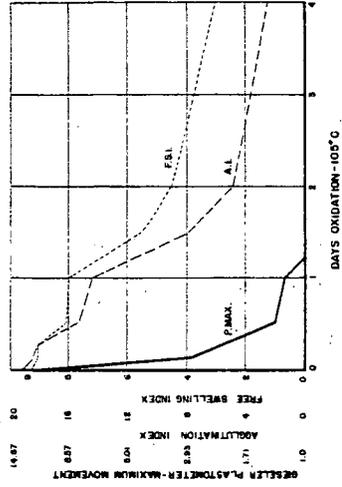


CHART IV
CHANGE IN MAXIMUM FLUIDITY IN STORAGE
EFFECTS OF COAL PARTICLE SIZE - SUNNYSIDE COAL

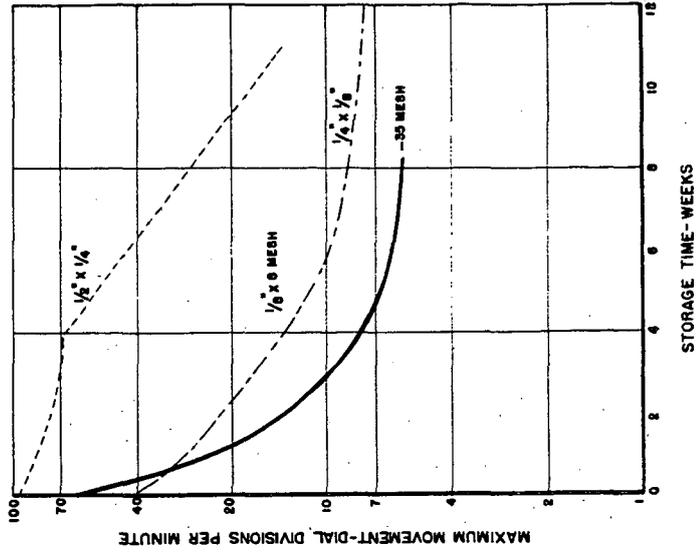


CHART III
CHANGE IN MAXIMUM FLUIDITY IN STORAGE
-35 MESH SUNNYSIDE COAL

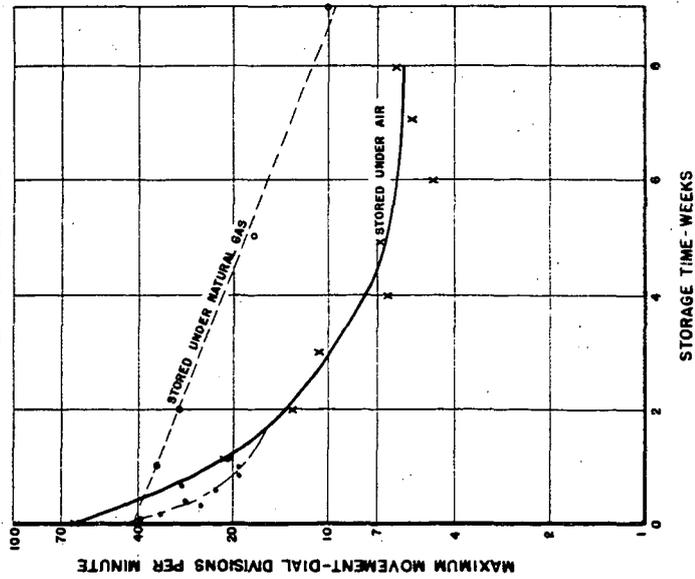


CHART VI
CHANGE IN MAXIMUM FLUIDITY WITH CHANGE IN COMPOSITION
SUNNYSIDE-MEDIUM VOLATILE COAL BLENDS

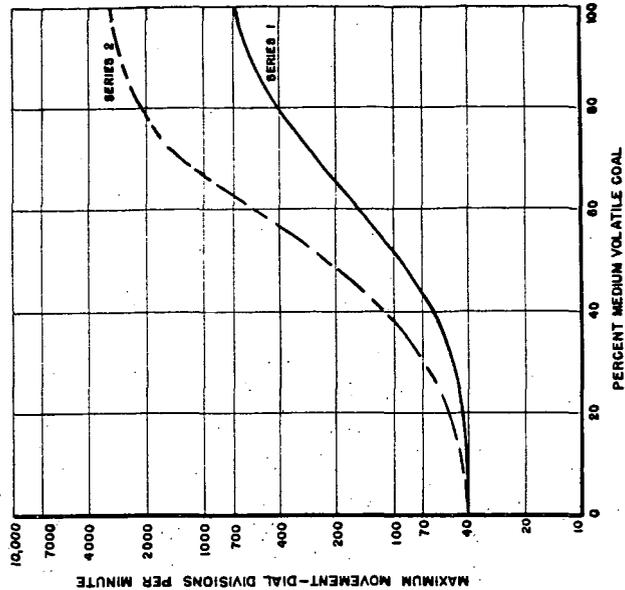
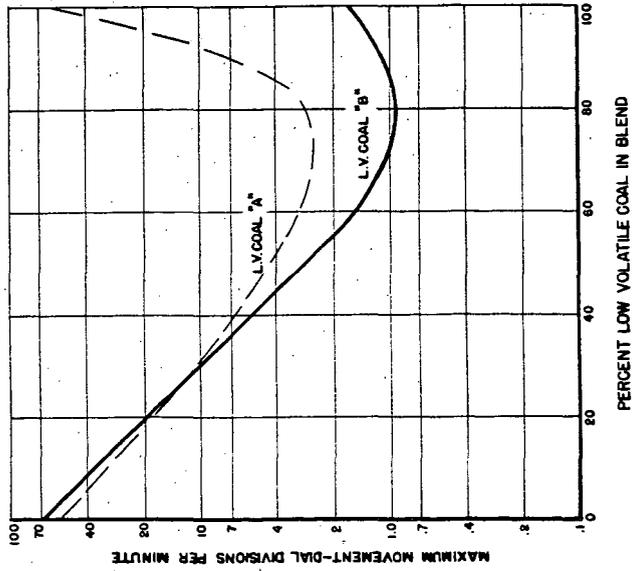
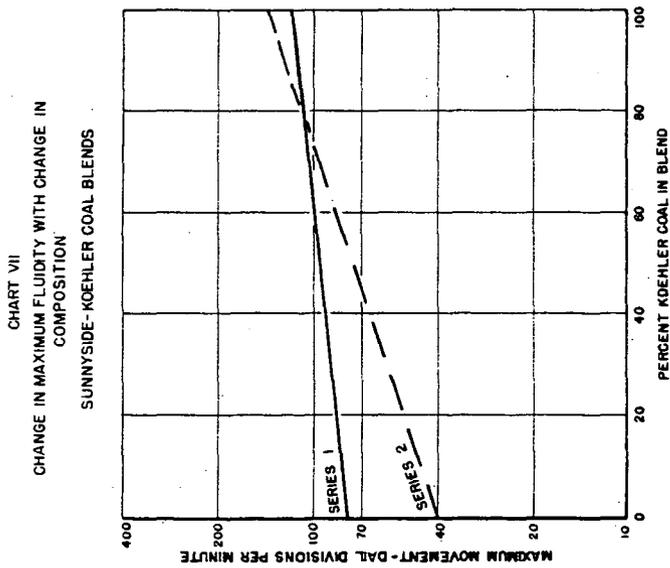
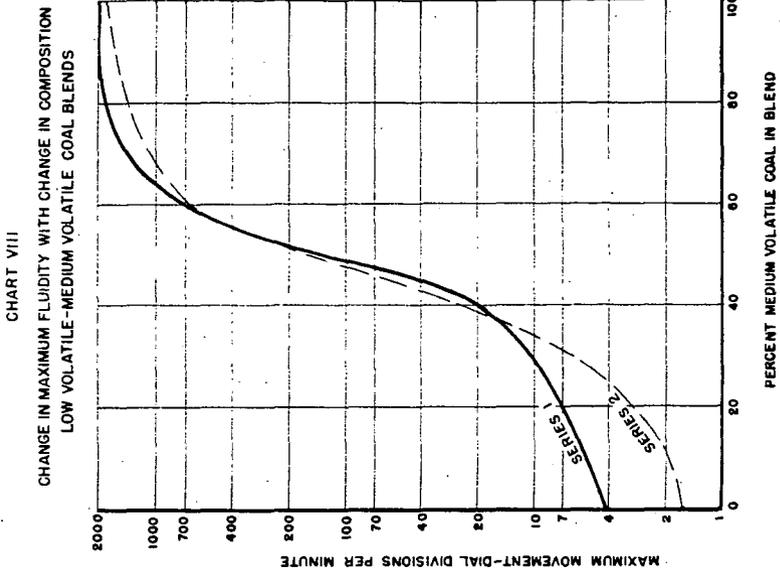


CHART V
CHANGE IN MAXIMUM FLUIDITY WITH CHANGE IN COMPOSITION
SUNNYSIDE-LOW VOLATILE COAL BLENDS





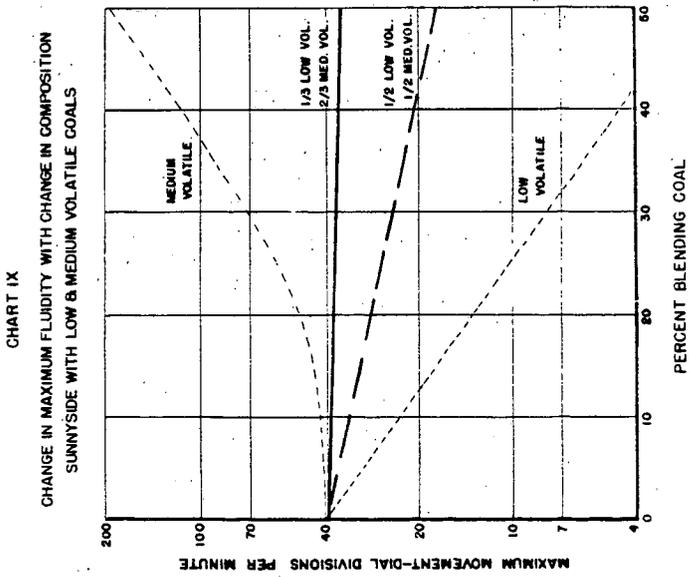
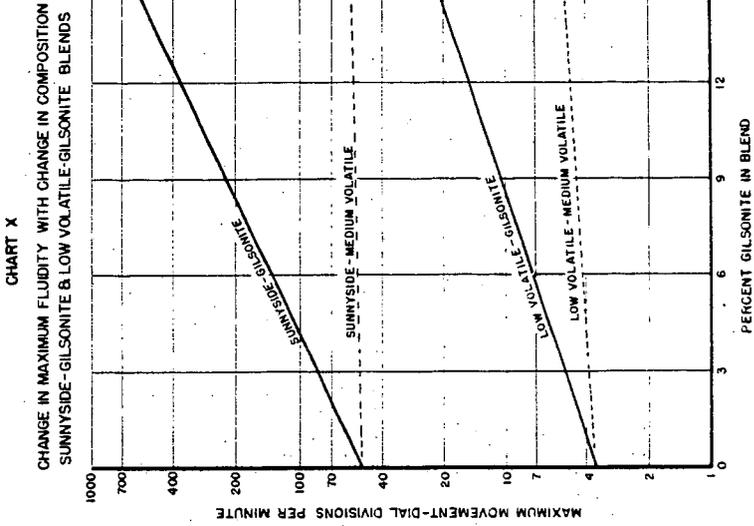


CHART XII
 CHANGE IN MAXIMUM FLUIDITY WITH CHANGE IN COMPOSITION
 SUNNYSIDE-PETROLEUM COKE & KOEHLER-PETROLEUM COKE BLENDS

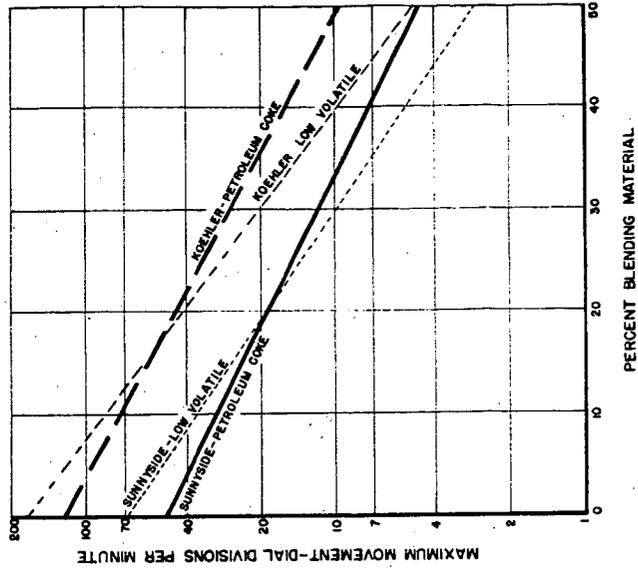
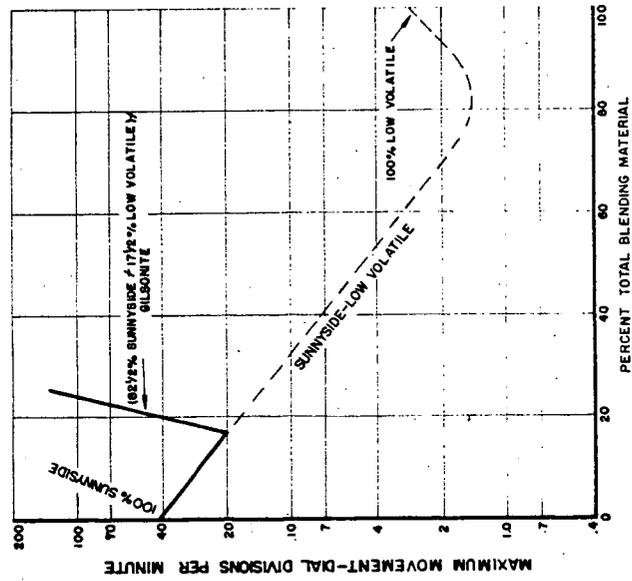


CHART XI
 CHANGE IN MAXIMUM FLUIDITY WITH CHANGE IN COMPOSITION
 SUNNYSIDE-LOW VOLATILE COALS WITH GILSONITE



THE RELATIONSHIP BETWEEN THE CHEMICAL AND PLASTIC PROPERTIES OF
ALABAMA MEDIUM-VOLATILE COALS AND THEIR CARBONIZATION BEHAVIOR

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Because coke quality is a significant factor in the efficient operation of blast furnaces, coke-plant operators are continually striving to produce the highest quality coke possible. No less important to the operators, however, are those characteristics of the coals or coal blends that influence the coking pressure and the ease with which each charge can be pushed from the oven. The difficulties in pushing coke from the ovens have been classified¹⁾* into two general categories: (1) those inherent in the particular coals or coal blends and (2) those due to plant practices. Apart from the difficulties attributable to plant practices, insufficient contraction of the coking charge at the end of the coking cycle is undoubtedly the most common reason for coke being hard to push from the oven. Insufficient contraction can be caused by either a lack of sufficient contraction inherent in the coal or coal blend, or by incomplete carbonization of the coal charge in the oven.

The expansion-contraction data obtained from a sole-heated oven is usually used to provide an indication of this aspect of the coking behavior of coal charges. As was pointed out recently,²⁾ a minimum contraction of 7 per cent in the sole-heated oven at the plant-operating bulk density is considered the limiting value for avoiding trouble caused by pushing difficulties. Other investigators^{3,4)} have shown that volume-change characteristics (or expansion-contraction properties) are related to the chemical and plastic properties of a wide range of coals. However, investigations⁵⁾ on coking-strength evaluations indicated that more workable relationships could be established if the correlations were confined to a narrower range of coals or coal blends. Accordingly, U. S. Steel's Applied Research Laboratory undertook a program to determine whether this concept was applicable in relating the volume-change characteristics of several Alabama medium-volatile coals to their chemical and plastic properties. Such a relationship would be extremely useful at U. S. Steel's Fairfield plant in Alabama.

Source and Identification of Coals

Thirty coal samples, 5 washed and 25 raw-mine samples, were examined for chemical, plastic, coking, and volume-change properties. The samples were obtained from the Pratt, American, and Mary Lee seams of the Warrior Coal Field in Alabama. The 5 washed samples were obtained by taking representative gross samples of about 250 pounds each of freshly mined coal from between 5 and 10 mine cars at the unloading station of each of 5 mines. These samples were crushed to pass 2-inch square openings and were experimentally washed to approximate the actual commercial preparation of the respective coals. The 25 raw samples were taken from different zones in the vertical face of the coal seam at specific site locations in the three seams, see Table I. Each sample was chosen by visual inspection of bright and dull coal in the seams, to give differences in coal type.

Experimental Procedures

In the preparation of the charges for carbonization tests, all coals were dried to a nominal moisture content of 1 per cent and pulverized to a 1/4-inch top size.

* See References.

The carbonization tests were conducted in the 30-pound test oven and in a sole-heated oven at the Applied Research Laboratory. Tests in the 30-pound test oven were conducted at the fast rate (1850 F) described in a previous publication.⁵⁾ The sole-heated oven used is similar in construction to the Bureau of Mines sole-heated oven⁶⁾ except that it is heated by four Globar heating elements and changes in the volume of the coal charge are continuously recorded by means of an inductance-bridge type instrument (also known as a differential transformer). The cokes produced in these ovens were tested according to the U. S. Steel Modified Tumbler Test procedure.⁵⁾

The proximate analyses and mineral-matter contents of the various samples are shown in Table I. For purposes of comparison, it was necessary to correct the as-measured volatile-matter contents of the coals to the Parr true-volatile-matter content basis.⁷⁾ The plastic properties of the coals are given in Table II. The volume change (expansion or contraction) in the sole-heated oven and the relative strengths of cokes from this oven are presented in Table III. The coking pressures and the strengths of the cokes from the 30-pound test oven are listed in Table IV.

In the sole-heated oven tests, significant differences in oven bulk densities were noted for various charges, with coals of lower ash content showing generally lower bulk densities than coals of higher ash content, as illustrated in Figure 1. Moreover, because the coals of higher ash content were more resistant to pulverization, the coarseness of the higher ash coals, expressed as the per cent retained on 8 mesh, was greater for these coals than for those of lower ash content, Figure 2. Therefore, because bulk density affects the as-measured volume change, these values were corrected⁸⁾ to a constant bulk density of 55 pounds per cubic foot and to 1 per cent of the as-charged coal retained on 4 mesh, to present the test results on a comparative basis. A reference bulk density of 55 pounds per cubic foot was used because this is approximately the highest level that can be attained with air-dried coal in commercial practice. Bulk density also influences coking pressure, but at the lower pressures encountered with these coals the effect is small.

Chemical and Plastic Properties of Coals

Most of these coals are classified as medium-volatile in rank. A few of the raw mine samples and the Pratt seam, Mine A, washed samples were on the borderline between medium-volatile and high-volatile A in the American Society for Testing Material (ASTM) classification.⁹⁾ The ash content of the raw samples ranged from about 3 to almost 26 per cent, and the sulfur contents from 0.6 to 5.6 per cent, Table I. The ash contents of the washed coals ranged between 7 and 11.7 per cent, and the sulfur contents between 0.8 and 1.7 per cent. The Pratt-seam coals showed the highest sulfur contents.

The plastic properties of these coals ranged from medium to high fluidity, Table II. When the sole-heated-oven volume-change results, Table III, were plotted against the Gieseler maximum fluidity, two principal behavior groups were evident, Figure 3. All coals having maximum fluidities above 10,000 dial divisions per minute (ddpm) were contracting within a range from about 5 to 30 per cent. Those coals having maximum fluidities below 10,000 ddpm ranged from mildly contracting, through neutral, to moderately expanding at about 2500 ddpm fluidity. The raw and washed Pratt Seam samples from Mine A; the raw American Seam, Site No. 2 samples; the raw Mary Lee Seam, Site No. 1 samples; and the washed Pratt Seam, Mine B samples all showed fluidities above 10,000 ddpm. The remaining samples showed lower fluidities. Therefore, the differentiation of coals observed as a result of the Gieseler Plastometer fluidity measurements indicates that the plastometer test may be useful in distinguishing between volume-change behavior groups in these coals. These results are in general agreement with the relationship shown by Naugle, Davis, and Wilson¹⁰⁾ for coals ranging from low volatile to high-volatile A.

Volume Change

The volume changes of the 30 samples after carbonization in the sole-heated oven were found to correlate with (1) volatile-matter contents for samples containing similar ash contents and (2) with ash contents for samples with similar volatile matter contents.

The volume change of each of the 5 washed coals used in this program, along with selected Bureau of Mines data¹⁰⁾ is plotted in Figure 4 against the corresponding volatile-matter and ash contents on the dry basis. For the 5 Pratt-seam coals (Mines A, B, and C) and 1 American-seam coal the volume change varied linearly from slightly expanding to strongly contracting as the volatile matter increased for ash contents between about 6 to 9 per cent. This same relationship is also evident for the 3 Mary Lee seam coals, which had a higher level of ash content of 11 to 12 per cent. In contrast to the washed coals, the raw Pratt-seam coals from Mine A, taken from the same site in the mine, and the washed sample from this same mine (all plotted in Figure 5) showed contraction varying linearly with ash content when true volatile-matter contents ranged between 30.8 and 31.8 per cent. These same general relationships were evident for the other coals used in this program except that the individual effects of volatile matter and ash were masked by one another.

General relationships within narrow ranges of volatile matter, similar to those noted above, were reported by Brown³⁾ for Pennsylvania and West Virginia coals; for these it was also shown that separate relationships existed between the expansion and the volatile matter of two low-volatile coals. Later work by Naugle, Davis, and Wilson⁴⁾ also showed this same type of relationship, except that expansion was plotted against the dry, mineral-matter-free fixed-carbon contents of the coals. The fixed-carbon content, calculated on the dry, mineral-matter-free basis should give a similar relationship to that with the true volatile-matter content, because the fixed-carbon content is simply 100 minus the volatile-matter content determined in the proximate analysis.

An examination of the volume-change behavior of the various raw samples in terms of their site and zone locations in the mine provided the following results. In the Pratt Seam Mine A samples, contraction decreased from top to bottom of the seam as the ash contents of the zone samples decreased from top to bottom. These relationships are shown in Figure 6. In the American seam samples the volume change of the Site No. 1 zone samples varied from strongly expanding at the top of the seam, to mildly contracting about one-third down from the top of the seam, and back to moderately expanding in the lower two thirds of the seam, Figure 7. Although the ash contents of these samples were relatively low, the mildly contracting zone had the highest ash content. In the Site No. 2 zone samples from this mine, the top and bottom zones were strongly contracting (about 17 to 20%), whereas the two middle zones were only moderately contracting (about 9%). The ash contents of these samples, all below 8 per cent, more than doubled between top and bottom.

In the Mary Lee Seam samples, the Site No. 1 zone samples changed from mildly contracting at the top of the seam to strongly contracting between about one-fourth and one-half way down from the top, dropped to mildly contracting about three-fourths of the distance from the top, and increased again to strongly contracting at the bottom, Figure 8. The Site No. 2 samples were almost neutral at a bulk density of 55 pounds per cubic foot in the sole-heated oven. The volume change varied from incipiently contracting at the top of the seam to mildly expanding between the top and about one-quarter of the way down from the top of the seam, to mildly contracting between one-quarter and one-third of the way down, to incipiently expanding between two-thirds of the way down from the top and to mildly contracting at the bottom.

Coke Strength

Because there was insufficient coal in each sample for carbonization tests in the Laboratory's 500-pound oven, an attempt was made to evaluate the strengths of the cokes from the sole-heated oven, Table III, and those of the corresponding cokes from the 30-pound test oven, Table IV. These relative strengths were plotted, Figure 9, in terms of the Modified Tumbler Index. The correlation coefficient for the relationship shown in Figure 9 was 0.94. In general, the cokes from the two ovens maintained similar relative positions: coals that produced strong cokes in the one oven also produced strong cokes in the other oven. Because of the wide differences in coking rates in the two ovens, the relationship that is valid for these coals cannot be expected to be generally applicable to other coals, especially to less strongly coking coals.

The influence of the ash contents of the raw and washed coals on the strengths of the test-oven cokes can be distinguished when the data are examined critically, but the full effect is masked and is difficult to establish conclusively because of the interrelationship between the ash content and the degree of pulverization of the various charges, shown in Figure 2. In the present tests, the larger average-particle size of some coals, particularly those in which the inert-type materials present are resistant to pulverization, resulted in more abradable cokes with lower Modified Tumbler Indexes than those produced from coals of smaller particle size, see Tables III and IV. Other coals of relatively high ash content produced cokes of strengths equivalent to or greater than cokes from coals of lower ash content, apparently because the inert materials present in the former were reduced to smaller particle size than those in the latter. When coals of similar ash content, either high or low, showed marked differences in degree of pulverization, those of coarser size-consist usually yielded test-oven cokes of lower strength.

Summary

The chief results of this investigation of the possible correlations existing between the carbonization behavior of these coals and their chemical and plastic properties were as follows:

(1) All coals, both raw and washed, can be classified in two broad fluidity and volume-change groups: (a) those showing fluidities above 10,000 ddpm, all of which were contracting, and (b) those below 10,000 ddpm, but above approximately 2500 ddpm, which were mildly contracting, neutral, or moderately expanding. Thus, measurement of fluidity in the Gieseler plastometer may be a means of classifying similar coals into at least two broad groups, according to the relative volume change observed in the sole-heated oven.

(2) For ash contents within narrow limits and for the range of volatile-matter contents determined in these coals, volume change in the sole-heated oven showed a linear change with dry-basis volatile-matter content. Likewise, for coals of a narrow range of true volatile-matter contents, contraction increased with increasing ash content. Consequently, it should be possible to correlate the volume change of these coals with their chemical properties. However, because variations existed in the raw and washed coals from the various seams used in this investigation, and are known to exist in practice, it is natural to expect variations in both the volatile-matter and the ash contents of these coals. It would appear, therefore, that a useful method of estimating the comparative pushing characteristics of these coals from an examination of their chemical properties should result from the use of the relative volume change obtained from a multiple correlation with corrected ash and volatile-matter contents.

(3) A significant correlation existed between the Modified Tumbler Indexes of the cokes produced in the sole-heated oven and those produced in the 30-pound test oven.

(4) The influence of ash content on coke strength was evident. However, the separate and full effect of ash content was masked by particle size, which has an important influence on coke strength, because increased ash content in the samples tested also indicated an increased amount of coarse, pulverization-resistant, inert-type, intermediate-gravity materials in the coal. The practical significance of this relationship is probably limited because the washed coals used in practice have ash contents that may not vary enough to appreciably affect the coke strength.

References

1. Gayle, J. B.; Eddy, W. H.; Brooks, J. A., "Studies of Coal Expansion," Chemical Engineering and Manufactured Gas Production Conference, Operation Section, American Gas Association, May 24 - 26, 1954, Pittsburgh, Pa., CEP-54-5.
2. Brisse, A. H., "Determination of Coke Oven Productivity From Coal Charge Characteristics," Blast Furnace and Steel Plant, Vol. 47, No. 4, April, 1959, pp. 376 - 383.
3. Brown, W. T., "Coal Expansion," Blast Furnace and Steel Plant, Vol. 30, January 1942, pp. 67 - 71, 219 - 23, 226.
4. Naugle, B. W.; Davis, J. D.; and Wilson, J. E., "Precision of Expansion Tests on Coal," Industrial and Engineering Chemistry, Vol. 43, December, 1951, pp. 2916 - 2922.
5. Price, J. G.; Shoenberger, R. W.; and Perlic, B., "Use of a Thirty-Pound Test Oven for Rapidly Assaying the Coking Strength of Coals," Blast Furnace, Coke Oven and Raw Materials Committee Proceedings, AIME (1958) Vol. 17, pp. 212 - 222.
6. Brown, W. T.; Coal Expansion: Proceedings American Gas Association, 1938, pp. 640 - 679.
7. Parr, S. W., Fuel, Gas, Water and Lubricants, 4th Edition, McGraw-Hill Book Co., New York, N. Y., pp. 49 - 55.
8. Naugle, B. W.; Wilson, J. E.; and Smith, F. W.; "Expansion of Coal in Sole-Heated Oven," U. S. Bureau of Mines, Report of Investigation 5295, January, 1957.
9. ASTM Designation: D388-38, Standard Specification for Classification of Coals by Rank.
10. Wilson, J. E.; Naugle, B. W.; and Wolfson, D. E.; "Expanding Properties of American Coals Tested in a Sole-Heated Oven," U. S. Bureau of Mines, Report of Investigation 5537, 1959.

Table I

Proximate Analyses and Calculated Mineral-Matter Contents (Dry Basis)

<u>Coal</u>	<u>Proximate Analysis,</u> <u>wt %</u>		<u>Ash</u>	<u>Sulfur,</u> <u>wt %</u>	<u>Calculated</u> <u>Mineral</u> <u>Matter, wt %</u>	
	<u>Volatile</u> <u>Matter</u>	<u>Fixed</u> <u>Carbon</u>				
<u>American Seam</u>						
Washed		26.7	65.8	7.5	0.9	8.6
Site No. 1	Zone 1	23.6	72.7	3.7	0.9	4.5
(Raw)	Zone 2	23.9	73.0	3.1	0.8	3.7
	Zone 3	22.9	66.4	10.7	1.0	12.1
	Zone 4	23.3	71.6	5.2	0.8	6.1
	Zone 5	24.6	69.8	5.6	1.1	6.7
Site No. 2	Zone 1	30.4	66.3	3.3	0.6	3.9
(Raw)	Zone 2	29.8	64.8	5.4	0.7	6.2
	Zone 3	30.3	65.2	4.5	0.6	5.2
	Zone 4	29.7	63.3	7.0	0.7	8.0
	Full Seam	30.0	63.5	6.5	0.6	7.4
<u>Pratt Seam, Mine A</u>						
Washed		30.2	62.3	7.5	1.7	8.3
Site No. 1	Zone 1	28.6	54.9	16.5	5.6	20.9
(Raw)	Zone 2	28.9	61.9	9.2	1.7	10.9
	Zone 3	30.4	66.2	3.4	1.7	4.6
	Zone 4	30.4	65.2	4.4	1.9	5.8
	Full Seam	27.5	57.6	14.9	2.1	17.3
<u>Mary Lee Seam</u>						
Washed		27.6	60.7	11.7	0.8	13.1
Site No. 1	Zone 1	29.0	58.6	12.4	0.7	13.8
(Raw)	Zone 2	28.3	54.0	17.7	1.1	19.7
	Zone 3	27.0	47.4	25.6	1.1	28.3
	Zone 4	29.5	61.3	9.2	0.6	10.3
	Zone 5	28.4	52.8	18.8	0.6	20.6
Site No. 2	Zone 1	24.8	59.1	16.1	1.8	18.4
(Raw)	Zone 2	24.9	60.8	14.3	0.6	15.8
	Zone 3	25.2	57.0	17.8	0.9	19.7
	Zone 4	25.1	62.6	12.3	0.6	13.6
	Zone 5	27.2	65.8	7.1	0.7	8.1
<u>Pratt Seam, Mine B</u>						
Washed		26.1	67.0	7.0	1.3	8.3
<u>Pratt Seam, Mine C</u>						
Washed		28.0	63.6	8.4	1.7	10.0

Table II
Plastic Properties

Coal	Gieseler*		Plastic Range, C			
			Dial Divisions		Softening	Solidification
			Per Min	At C	Temperature	Temperature
<u>American Seam</u>						
Washed		7,400	449	380	515	
Site No. 1 (Raw)	Zone 1	4,000	458	391	504	
	Zone 2	3,400	454	385	503	
	Zone 3	3,800	459	390	502	
	Zone 4	2,800	476	390	506	
	Zone 5	2,700	450	385	505	
Site No. 2 (Raw)	Zone 1	18,500	438	376	494	
	Zone 2	12,000	439	371	482	
	Zone 3	18,500	437	371	493	
	Zone 4	16,500	434	375	-	
	Full Seam	18,500	442	373	494	
<u>Pratt Seam, Mine A</u>						
Washed		16,500	436	348	496	
Site No. 1 (Raw)	Zone 1	14,000	432	350	489	
	Zone 2	20,000	437	356	503	
	Zone 3	17,500	432	355	486	
	Zone 4	20,500	434	354	497	
	Full Seam	19,000	437	357	500	
<u>Mary Lee Seam</u>						
Washed		9,500	442	362	495	
Site No. 1 (Raw)	Zone 1	12,500	448	371	490	
	Zone 2	15,000	441	370	491	
	Zone 3	21,500	442	370	484	
	Zone 4	12,000	447	361	490	
	Zone 5	14,000	436	363	489	
Site No. 2 (Raw)	Zone 1	3,500	444	367	499	
	Zone 2	3,500	458	369	500	
	Zone 3	5,000	446	364	498	
	Zone 4	7,000	444	371	494	
	Zone 5	8,500	446	362	497	
<u>Pratt Seam, Mine B</u>						
Washed		6,000	449	391	502	
<u>Pratt Seam, Mine C</u>						
Washed		13,000	439	368	500	

* No washer was used on the Gieseler plastometer retort to prevent swelling.

Table III

Sole-Heated Oven Test Results

<u>Coal</u>	<u>Volume Change</u> (as Measured), %		<u>Oven</u> <u>Bulk Density,*</u> <u>lb/cu ft</u>	<u>Corrected</u> <u>Volume Change,</u> % (at 55 lb/cu ft)	<u>Modified Tumbler</u> <u>Index of Coke</u> <u>Cum % + 3/4 in.</u>
	<u>Maximum</u>	<u>Final</u>			
<u>American Seam</u>					
Washed		-4.1	55	-2.9	75
Site No. 1	Zone 1	-3.8	48	+11.3	74
(Raw)	Zone 2	+1.5	51	+7.9	77
	Zone 3	-7.8	53	-2.7	56
	Zone 4	-3.6	51	+7.8	75
	Zone 5	+1.0	52	+5.9	69
Site No. 2	Zone 1	-20.6	48	-17.1	62
(Raw)	Zone 2	-13.5	48	-9.1	58
	Zone 3	-18.7	50	-8.9	66
	Zone 4	-27.8	51	-20.7	43
	Full Seam	-18.8	50	-9.0	57
<u>Pratt Seam, Mine A</u>					
Washed		-25.2	52	-20.1	68
Site No. 1	Zone 1	-32.2	56	-31.9	55
(Raw)	Zone 2	-27.7	52	-22.8	36
	Zone 3	-20.6	52	-14.7	72
	Zone 4	-15.4	52	-10.3	72
	Full Seam	-30.0	56	-29.9	14
<u>Mary Lee Seam</u>					
Washed		+0.3	54	0.0	73
Site No. 1	Zone 1	-9.1	53	-3.4	58
(Raw)	Zone 2	-20.6	56	-19.9	24
	Zone 3	-21.4	58	-21.8	43
	Zone 4	-12.1	51	-4.4	67
	Zone 5	-24.3	53	-20.0	18
Site No. 2	Zone 1	-5.1	53	-0.6	74
(Raw)	Zone 2	+1.3	53	+3.5	70
	Zone 3	-8.0	54	-4.9	46
	Zone 4	-3.4	53	+0.8	69
	Zone 5	-7.2	53	-3.2	69
<u>Pratt Seam, Mine B</u>					
Washed		+3.1	55	+1.6	75
<u>Pratt Seam, Mine C</u>					
Washed		-9.0	55	-8.1	72

* One % moisture content.

Table IV

Coking-Pressure and Modified-Tumbler-Index Results
(Conducted in 30-Pound Test Oven)

<u>Coal</u>	<u>Coking Pressure, psi</u>	<u>Oven Bulk Density,* lb/cu ft</u>	<u>Modified Tumbler Index cum % ● 3/4 in.</u>
<u>American Seam</u>			
Washed	1.0	47	68
Site No. 1 Zone 1	1.0	44	71
(Raw) Zone 2	1.2	46	66
Zone 3	1.6	46	54
Zone 4	1.9	45	64
Zone 5	0.9	45	65
Site No. 2 Zone 1	0.6	45	58
(Raw) Zone 2	1.0	45	62
Zone 3	0.7	48	61
Zone 4	0.8	44	59
Full Seam	0.6	44	58
<u>Pratt Seam, Mine A</u>			
Washed	1.5	48	65
Site No. 1 Zone 1	1.2	47	62
(Raw) Zone 2	1.0	46	50
Zone 3	-	-	-
Zone 4	1.6	45	62
Full Seam	0.9	48	24
<u>Mary Lee Seam</u>			
Washed	2.1	49	70
Site No. 1 Zone 1	1.0	45	58
(Raw) Zone 2	1.0	48	41
Zone 3	1.4	50	50
Zone 4	1.3	46	61
Zone 5	1.4	47	26
Site No. 2 Zone 1	2.8	48	72
(Raw) Zone 2	2.3	46	68
Zone 3	1.0	49	50
Zone 4	-	-	-
Zone 5	1.0	45	70
<u>Pratt Seam, Mine B</u>			
Washed	-	-	-
<u>Pratt Seam, Mine C</u>			
Washed	1.0	47	65

* One % moisture content.

Figure No. 1

EFFECT OF ASH CONTENT ON BULK DENSITY

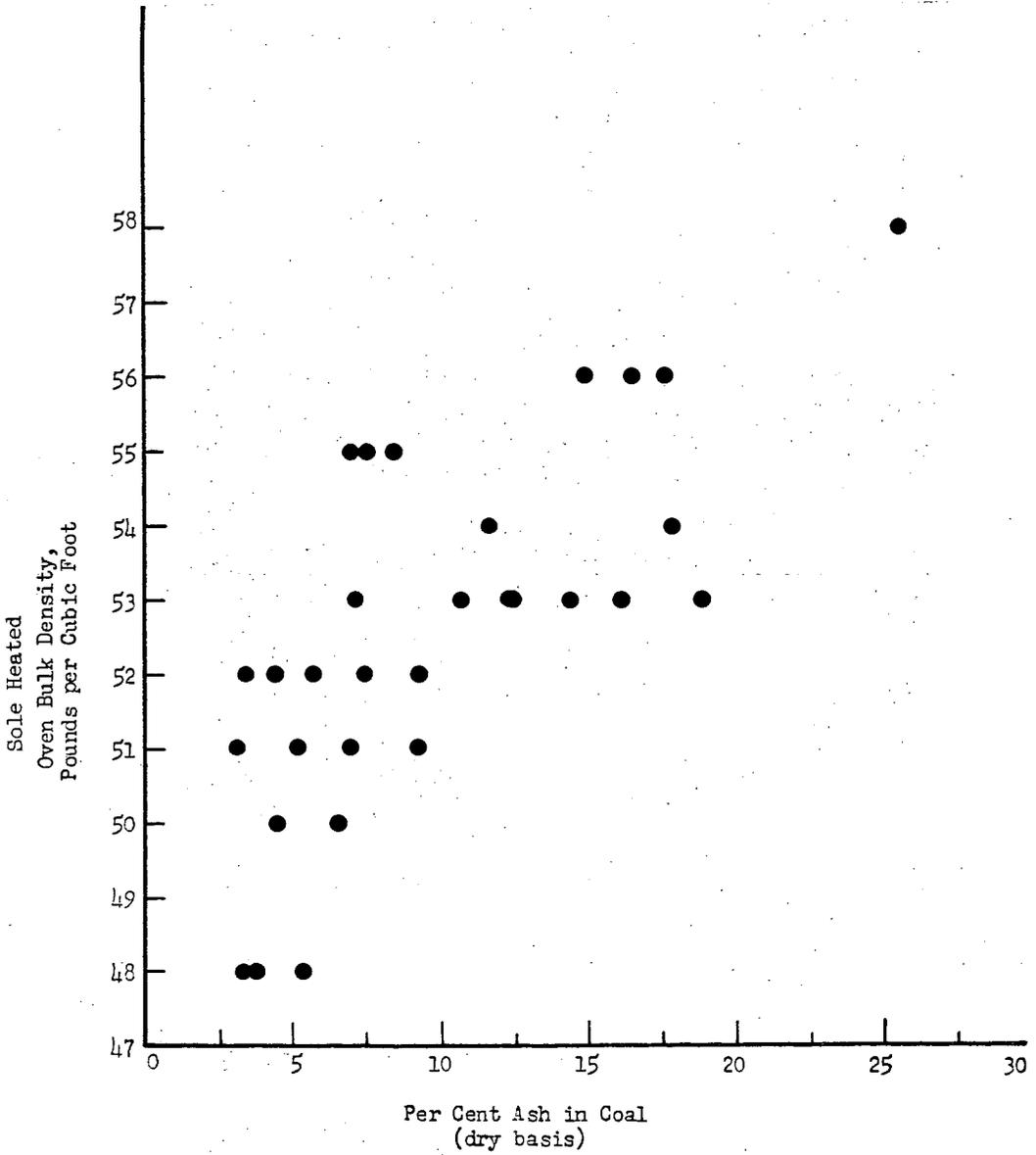


Figure No. 2

EFFECT OF ASH CONTENT ON PULVERIZATION (PER CENT RETAINED ON 8-MESH)

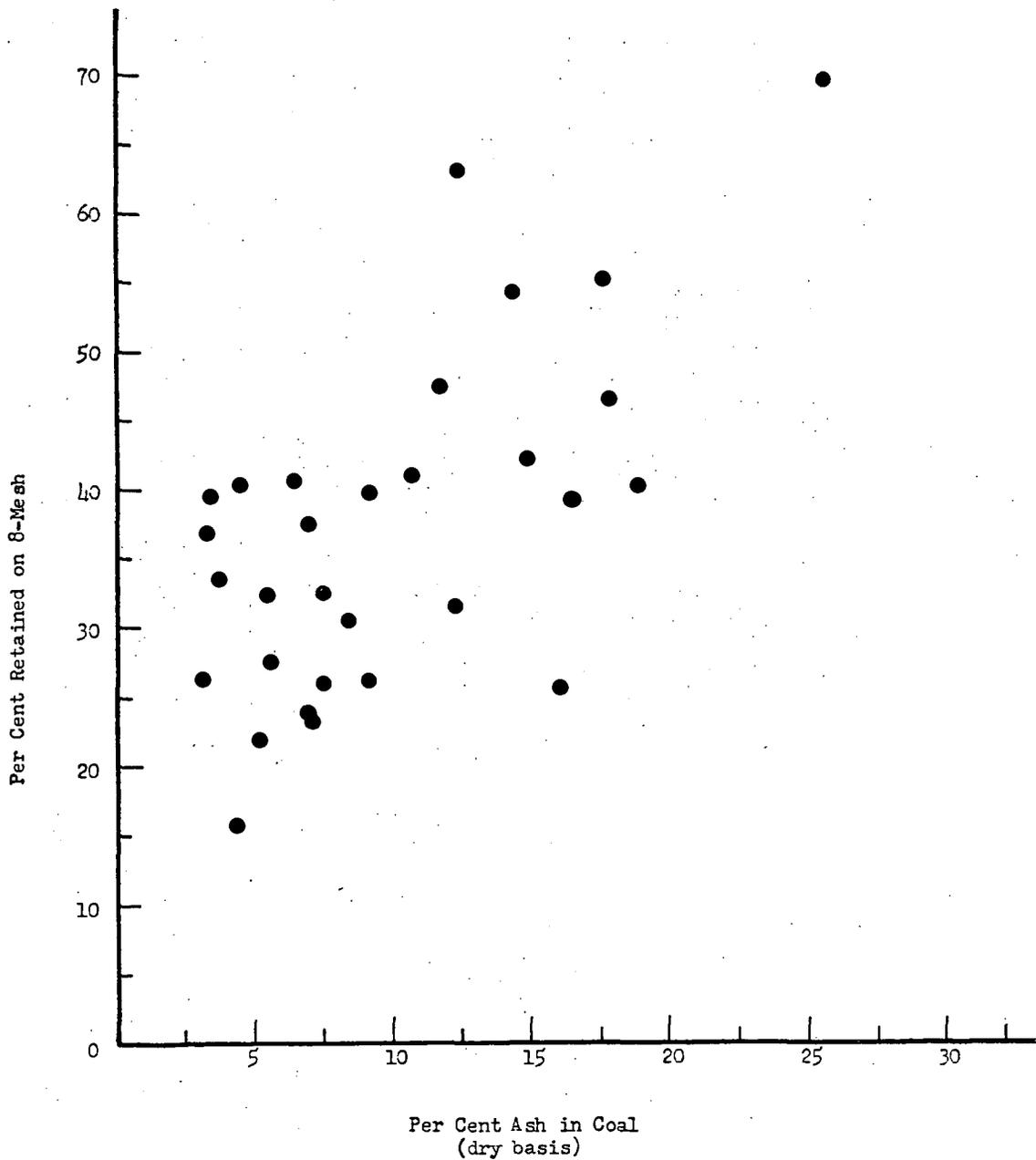
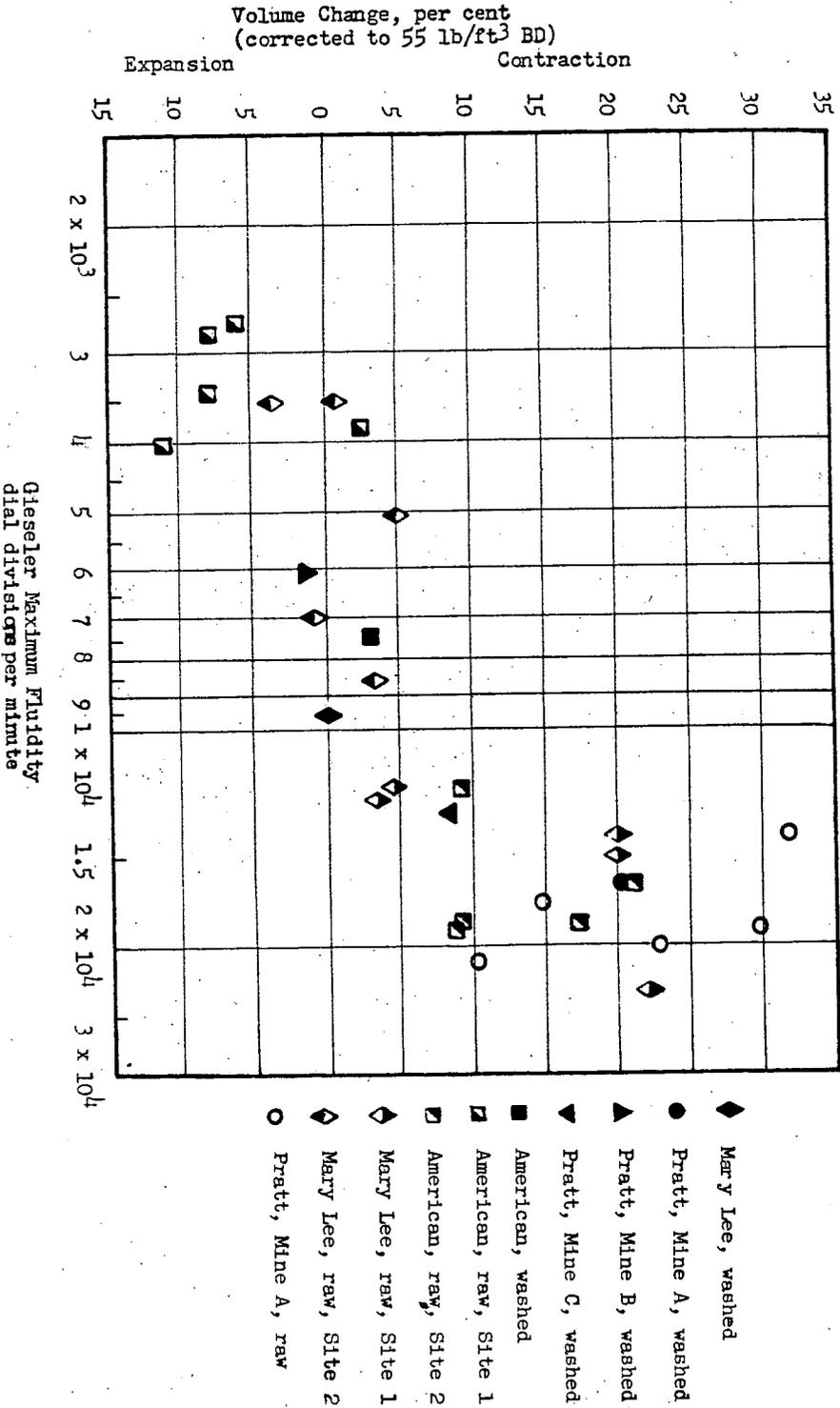


Figure No. 3

RELATIONSHIP OF VOLUME CHANGE IN THE SOLE-HEATED OVEN AND MAXIMUM FLUIDITY



RELATIONSHIP OF VOLUME CHANGE IN THE SOLE-HEATED OVEN TO VOLATILE MATTER AND ASH CONTENTS OF WASHED COALS

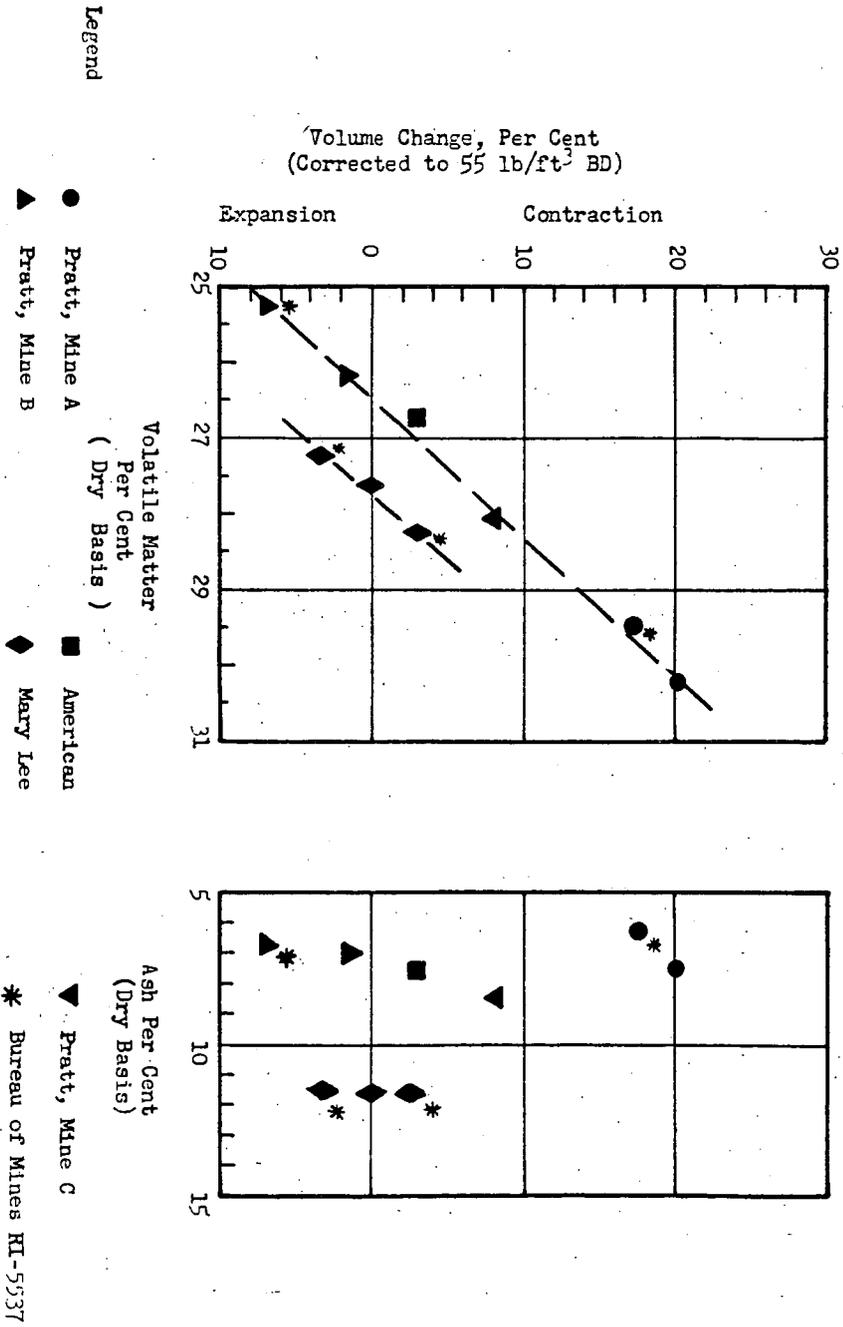


Figure No. 5

RELATIONSHIP OF CONTRACTION IN THE SOLE-HEATED OVEN TO ASH AND VOLATILE MATTER CONTENTS OF PRATT SEAM MINE A, COALS

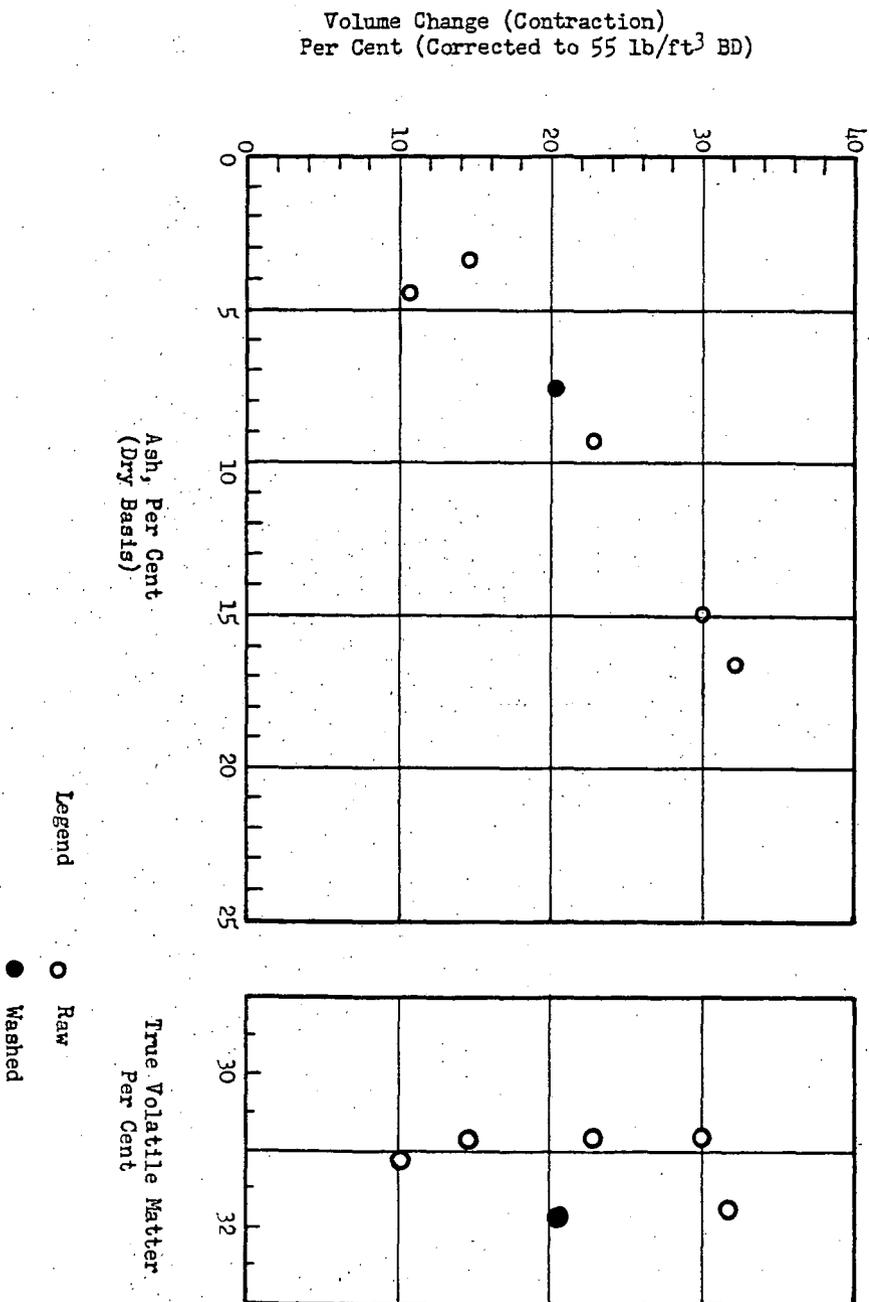


Figure No. 7

COMPARISON OF SITE AND ZONE SAMPLES FROM AMERICAN SEAM MINE

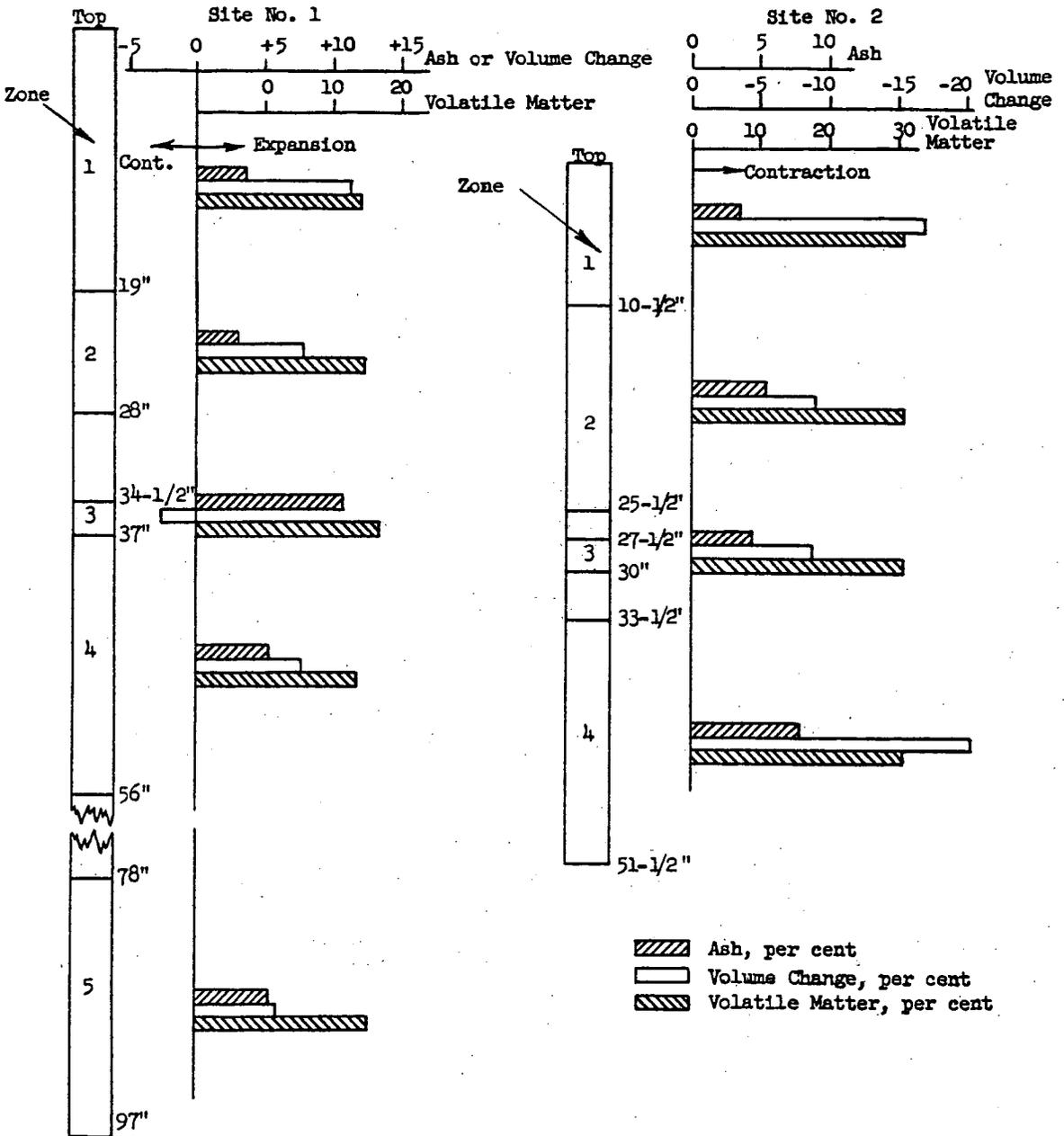


Figure No. 6

COMPARISON OF ZONE SAMPLES FROM THE PRATT SEAM, MINE A

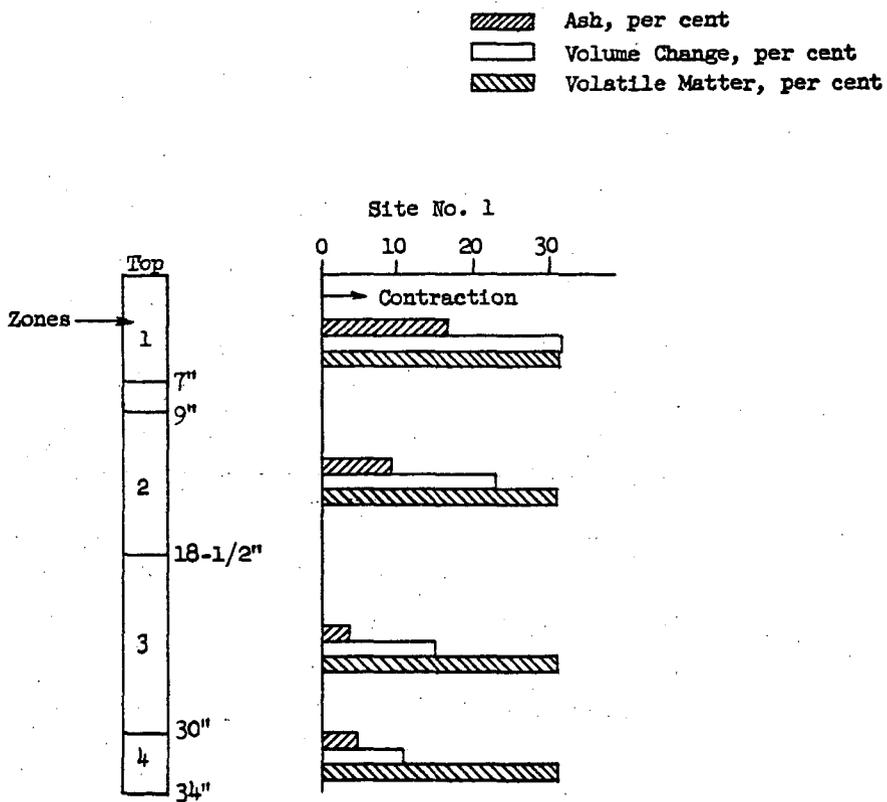


Figure No. 8

COMPARISON OF SITE AND ZONE SAMPLES FROM MARY LEE SEAM MINE

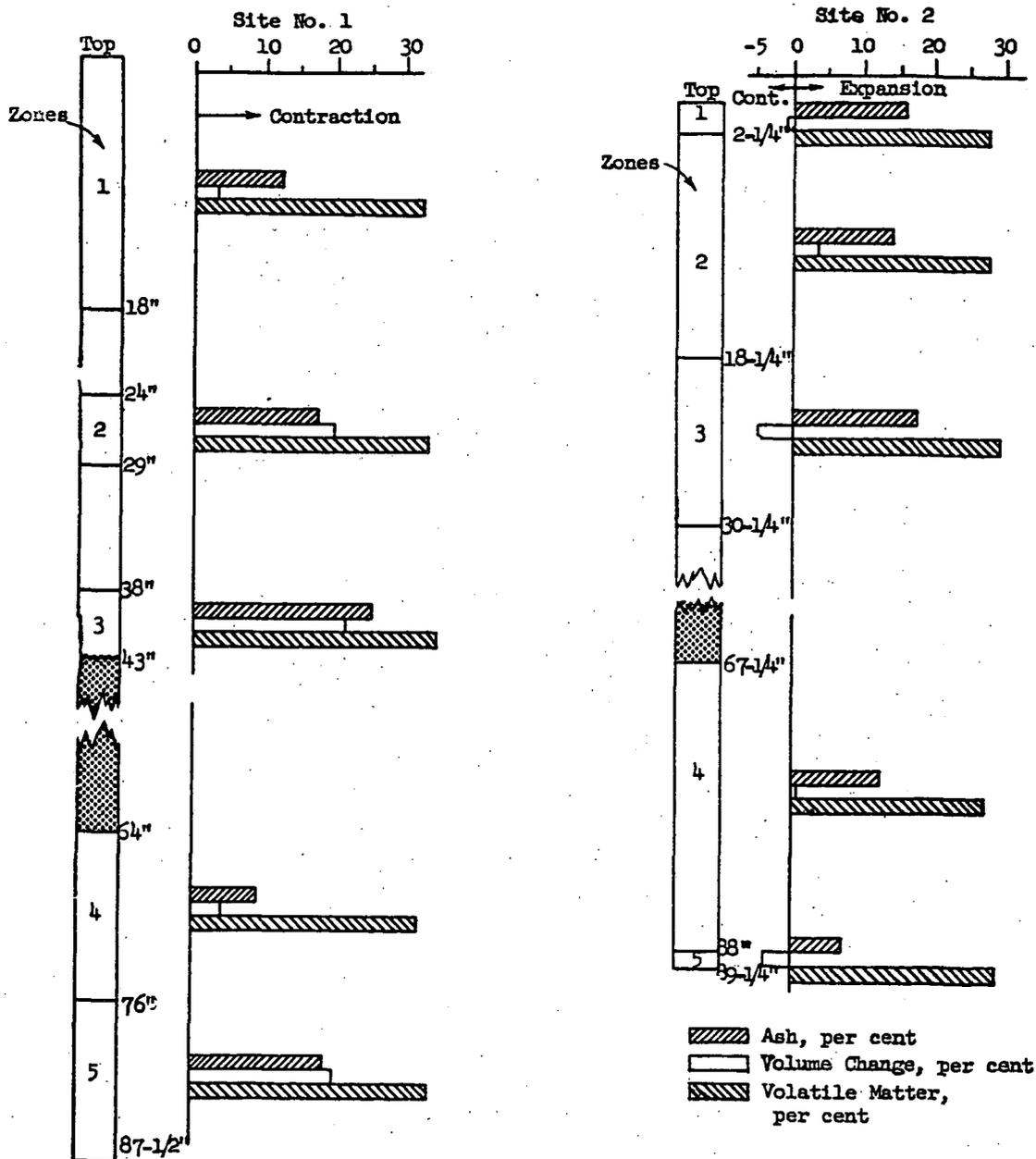
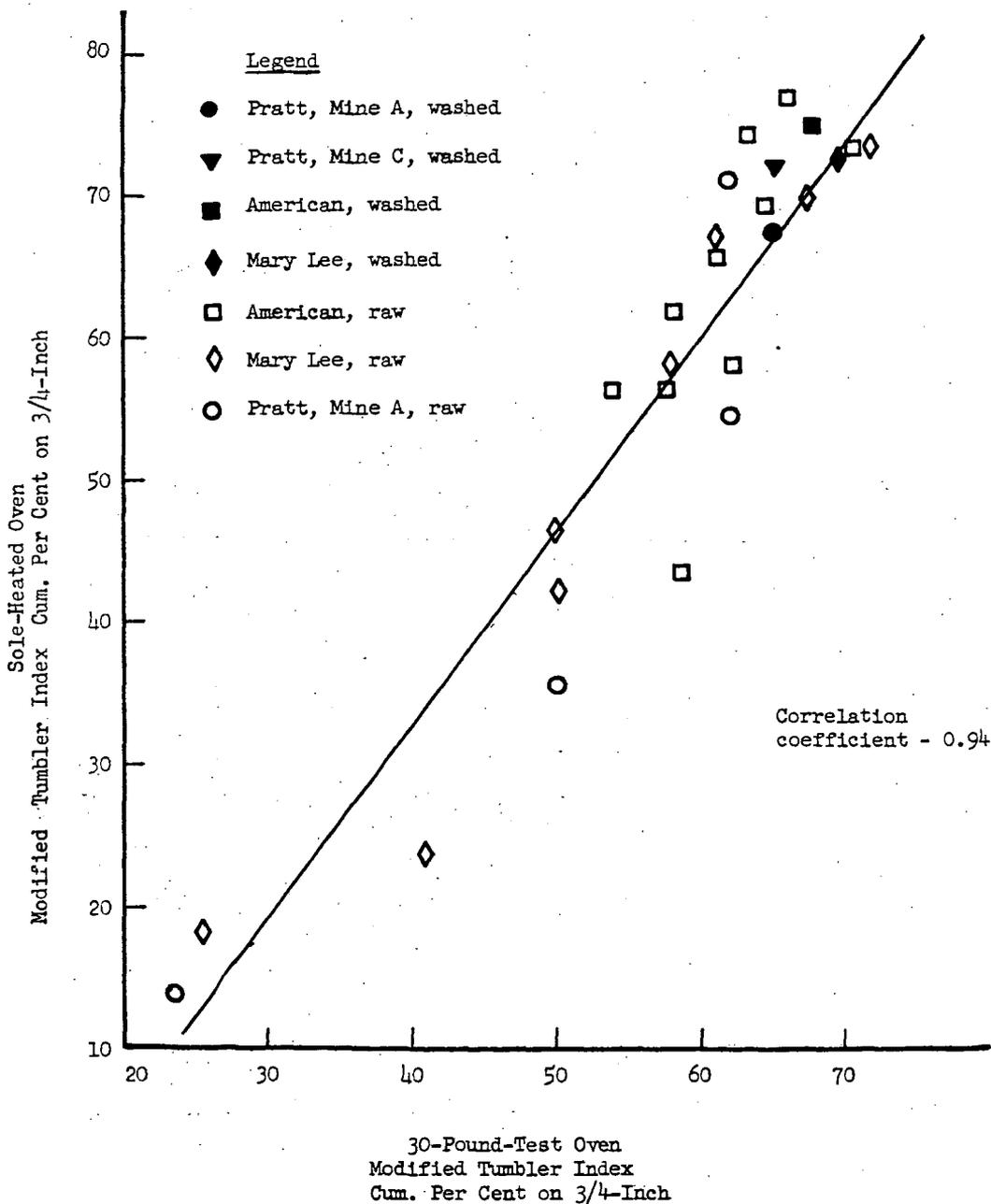


Figure No. 9

RELATIONSHIP BETWEEN 30 POUND TEST AND SOLE-HEATED OVENS 3/4-INCH
TUMBLER INDEXES



THEMAL CONDUCTIVITY OF CARBONACEOUS BRIQUETTES

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Introduction

The purpose of this work was to determine the effect of temperature and briquetting pressure on the thermal conductivity of briquettes made from bituminous coal, anthracite, and petroleum fluid coke. The briquettes were formed by pressing these materials with suitable binders.

In spite of the promising future for carbonized briquettes of these materials there is a dearth of fundamental and systematic data on their properties, such as thermal conductivity. There is a need particularly for thermal conductivity data which would be used for the proper design of briquette carbonizing retorts and briquette using furnaces. Consequently it was felt that any further study, such as covered by this paper, will go a long way towards upgrading this process from its present stage as an art to one based upon sound technological principles.

Briquetting

Coals having the properties shown in Table I were dried in an oven at 225°F and ground in a hammer mill through a 3/64" screen. The bituminous coal was then contacted with air at 550°F until it was adequately oxidized to reduce coking power as observed by the ASTM free swelling test. The petroleum coke also shown in Table I was dried and ground in a ball mill for 15 minutes.

The pulverized coals and coke were blended with suitable binders, i.e., coal with coal tar pitch and coke with coker bottoms. The properties of the binders which were chosen on the basis of practical considerations as to general availability and adequacy are shown in Table II.

The mix was then pressed in a hydraulic press to form a cylindrical briquette, 3 1/8" in diameter, and approximately 1" thick. Briquetting pressures of 1000, 3000, 4000 and 5000 psig were used to determine subsequently the effect of pressure on thermal conductivity.

Table I - Properties of Carbonaceous Materials

	<u>Anthracite</u>	<u>Bituminous</u>	<u>Coke</u>
Fixed C	80.1%	73.3%	86.8%
Ash	15.4%	6.5%	0.0%
Volatile	4.1%	19.5%	6.1%
Sulfur	0.4%	0.7%	6.1%

Table II - Properties of Binders

	<u>Pitch</u>	<u>Coker Bottoms</u>
Conradson C	45.8%	33.0%
Ultimate Analysis - Carbon	94.0%	84.0%
Hydrogen	5.1%	8.5%
Sulfur	0.6%	5.2%
Ash	0.2%	

*Present address: Chemical Construction Corporation, New York, New York

**Present address: Scientific Design Corporation, New York, New York

Carbonization

A liner was made to fit an ordinary laboratory muffle furnace in order to drive off the volatile matter and designed to carbonize the briquettes at various temperatures in an inert atmosphere of nitrogen. The nitrogen was preheated in the outer section of the liner before coming in contact with the briquettes. The furnace was heated at a rate of 3° to 5°F per minute for the petroleum coke and anthracite, while for the bituminous coal this heat-up rate was maintained up to 900°F and reduced to 1°F per minute above this temperature. Experience with these materials had shown that, if these rates were adhered to, cracking due to too rapid vaporization of the volatile matter and shrinkage of the briquettes could be avoided. The purpose of this procedure was to drive off the volatile matter so that it would not subsequently foul the conductivity cell and so that the dimensions of the test specimen could be stabilized and be accurately measured prior to the determination.

Thermal Conductivity

The thermal conductivity cell used in these determinations was based on that described by J. S. Finck (1, 2) modified to enable these determinations to be conducted in an atmosphere of nitrogen in order to prevent combustion of the samples. A cross-section diagram of the cell is shown in Figure I with an enlarged view showing the relative locations of the thermocouples, test heater, guards and sample in Figure II.

The test heater element was imbedded in a 3" diameter alundun plate and delivered 250 watts at 110 V. In the same plate was a guard heater 2½" wide, which formed a ring around the test heater. The bottom guard heater was separated from the other heaters by 2½" of diatomaceous earth. Atop each of these heating elements was an isothermal plate of 3/4" silicon carbide in which chromel alumel thermocouples were imbedded. The samples, which, after shrinkage during carbonizing, were slightly over 3" in diameter, were placed on the test heater isothermal plate and covered by an additional silicon carbide isothermal plate.

The housing was 22" in diameter by 23" overall height. The cell was sealed by a 14" flange cover in order to make it gas tight. A water cooler, to provide an adequate temperature differential, was attached to the flange cover.

The inside of the cell housing was lined with 5" of 2000°F castable insulation. The spaces between the cell components and the lining were filled with loose diatomaceous earth.

The heaters were controlled by means of powerstats, 45 amp, 110 V for the guard heaters and 7.5 amp, 110 V for the test heater. The power input to the test heater was measured by a 0-500 ma milliammeter and 0-50 V voltmeter at low power levels and 100/200 watt dual range wattmeter at higher levels. This wattmeter was accurate to ½ watt.

The actual temperature differential across the test piece was measured by stainless steel sheathed chromel alumel thermocouples at the points indicated in Figure II.

The formula for thermal conductivity in a steady state system is as follows:

$$K = \frac{Qs}{At}$$

where K = thermal conductivity
Q = quantity of heat transferred
s = thickness of sample

A = cross sectional area of heat transfer
 t = temperature differential across sample

In order for this equation to be valid certain conditions must be met. 1) The system must be in a state of thermal equilibrium. 2) The heat flow must be normal to the cross sectional area through the entire height of the sample. 3) All the heat generated in the test heater and only this heat must flow through the sample. 4) The temperature differential must be reasonably small. The guarded hot plate cell used in this study substantially satisfied the first three conditions, and minimized the ill effects of the fourth.

If the first condition is satisfied by maintaining the cell at constant temperature for a reasonably long period of time and the third condition is satisfied by maintaining the test, ring guard and bottom guard isothermal plates at essentially the same temperature, thus negating any lateral or downward thermal driving forces.

It can be shown by the following analysis that the second condition is also satisfied:

The heat generated in the test heater flows upward into the test sample except for a small amount which flows into the gap between the ring guard and test heaters and then flows upward. (See Figure II) By placing an isothermal plate on top of the test sample there is little tendency for heat migration toward point 3, since the temperature at points 3 and 4 are now essentially equal. With this top plate the heat flow will tend to diverge toward point 3 due to the difference in conductivity in the test sample and the insulating material above the ring guard plate and the resultant temperature difference at points 3 and 4. The same analysis would hold for the heat generated in the ring guard heater. An additional item which effects the direction of the flow of heat from the ring heater is the effect of heat losses. Heat losses from the ring will affect the flow of heat through the test sample if they become excessive. However, with adequate insulation the adverse effect of end losses can be kept within the limits of experimental accuracy.

The fourth condition must be somewhat satisfied because the conductivity is actually a mean value between the conductivity at hot and cold face temperatures. The use of this value assumes that a linear relationship exists between conductivity and temperature.

The values for thermal conductivity determined in this study are tabulated below in Table III and plotted in Figure III. In all cases, except were noted, the briquettes were pre-carbonized at approximately the same temperature as the conductivity determination.

TABLE III

Material	Briq. Press. PSIG	Mean Temp. °F	K BTU/HR-°F-FT ² /IN	Material	Briq. Press. PSIG	Mean Temp. °F	K BTU/HR-°F-FT ² /IN
Anthracite	1000	295	2.69	Coke	1000	285	4.10
	3000	287	2.51		2000	293	3.80
	5000	284	2.87		5000	309	3.92
	1000	890	4.09		4000	726	3.36
	3000	894	3.76		1000	888	2.97
	5000	916	3.80		3000	928	2.82
	5000	1271	5.36		3000	1277	2.56

Table III (Cont.)

Material	Briq. Press PSIG	Mean Temp. °F	K BTU/ HR-°F-FT ² / IN
Bituminous	1000	263	1.36
	3000	278	1.23
	5000	277	1.18
	3000	591	1.18
	1000	941	2.91
	3000	880	3.28
	5000	865	2.88
	5000	1234	5.61
	1000	1466	9.98
	1000*	475	7.10
	1000*	1116	8.0

*These briquettes were carbonized at 1600°F to show effect of temperatures.

A study by Batchelor et al (3) on briquettes formed from 25% Pittsburg seam bituminous coal, 63.5% low temperature char of this material, and 11.5% pitch is substantially in agreement with the results of the present study on bituminous coal briquettes. In both cases the material was pre-carbonized.

Table IV

Temp. °F	K-BTU/HR-°F-FT ² /IN	
	<u>Batchelor et al</u>	<u>Present Study</u>
300	1.8	1.2
600	1.8	1.2
900	2.4	3.0
1200	4.8	5.6
1500	9.6	10.0

The only other thermal conductivity data found there the results were determined at elevated temperatures is that of Terres (4) on pieces of English coking coals. The values are in reasonable agreement with the present study. The work of Beletzki (5) on pulverized coals is also in good agreement considering that his values were determined at room temperature and to be compared to the current study should include a temperature coefficient. This data together with that of Batchelor et al is shown in Figure IV.

An attempt to show the effect of temperature alone involved the measurement of conductivities on a briquette which had been carbonized at 1600°F. The conductivities of this briquette at lower temperatures were considerably higher than the conductivities of briquettes carbonized at the temperature of the conductivity determination. (See Figure V)

From the above discussion, it will be evident that the values for thermal conductivity determined in this study are values which include the effect of temperature and the effect of carbonization. For design considerations this is the most useful conductivity value.

An attempt was also made to determine the effect of briquetting pressure on the thermal conductivity. Samples briquetted at three different pressures were measured at 300°F and 900°F, and the variations found were well within the accuracy of the determination and therefore could not be attributed to differences in briquetting pressures.

As a result of these tests, it is felt that the conductivity of briquetted coals is dependent on the nature of the base material only slightly modified by the binding agent and independent of the pressure effects of briquetting. Other properties probably enter into the picture, such as particle size, but these were beyond the scope of this study.

It is possible to formulate equations for the conductivities of fluid coke and anthracite, within the range studied, but the equation for the curve for bituminous coal would require a factor related to the coking power of the coal. The equations for fluid coke and anthracite include only a temperature factor since the carbonaceous base in these cases has not undergone any substantial thermal change. If the study could have been continued to higher temperatures, the coefficient of thermal change would probably enter into these equations also.

These conclusions are quite reasonable since the bituminous coal undergoes fusion and loses its particulate nature on carbonizing, while the other materials remain particulate even at much higher temperatures then encountered in this study. They do, however, give up volatile matter consisting mostly of hydrogen indicating a destructive distillation which could probably affect the thermal conductivity.

The equations for the thermal conductivity of the briquetted material based on the curves shown in Figure III are as follows:

Anthracite

$$300-1200^{\circ}\text{F} \text{ -- } K = 2.30 + 1.0 \times 10^{-3}t + 9.0 \times 10^{-7}t^2$$

Bituminous Coal

$$\begin{aligned} 300^{\circ} - 600^{\circ}\text{F} & \text{ -- } K = 1.20 \\ 600^{\circ} - 900^{\circ}\text{F} & \text{ -- } K = 0.0049t - 1.59 \\ 900^{\circ} - 1200^{\circ}\text{F} & \text{ -- } K = 0.0087t - 5.01 \\ 1200^{\circ} - 1500^{\circ}\text{F} & \text{ -- } K = 0.0150t - 13.70 \end{aligned}$$

Fluid Coke

$$300-1200^{\circ}\text{F} \text{ -- } K = 4.32 - 1.42 \times 10^{-3}t$$

K is the thermal conductivity in BTU/HR-°F - FT²/IN
t is temperature in °F.

The average deviation about the mean value for these determinations is 5 to 8% by measurement. The absolute accuracy, as calculated from possible errors in the measurement of temperature and heat input, is in the order of 15 to 20%. This is well within the limits required for engineering work and owing to the very specialized nature of the briquettes and the high degree of variation found in the characteristics of coals, is about as accurate as would be required.

BIBLIOGRAPHY

1. Finck, J.L., Journal of Amer. Ceram. Society, Vol. 18, pp 6-12, 1935.
2. Finck, J. L., Journal of Amer. Ceram. Society, Vol. 20, pp 378-82, 1937.
3. Batchelor, J. D., Yavorsky, P. M., and Gorin, E., "Measurement of the Thermal Properties of Carbonaceous Materials". Paper presented before the Division of Gas and Fuel Chemistry, American Chemical Society, Chicago, Illinois Meeting September 7-12, 1958.
4. Terres, E., Proc. 2nd. Int. Conf. Bit. Coals, Vol. 2, pp 679-83, 1928.
5. Beletski, A. F., Coke and Chemistry, USSR, Vol. 10, No. 3, pp 24-46, 1940.
6. Clendenin, J. D., Barclay, K. M., Donald, H. J., Gillmore, D. W., and Wright, C. C., "Thermal and Electrical Properties of Anthracite and Bituminous Coals", Pennsylvania State College, Minerals Industries Experimental Station Technical Paper 160, 1950.
7. ASTM Standards 1952, "Standard Method of Test For Thermal Conductivity Of Materials By Means Of The Guarded Hot-Plate", A.S.T.M. Designation C 177-45.

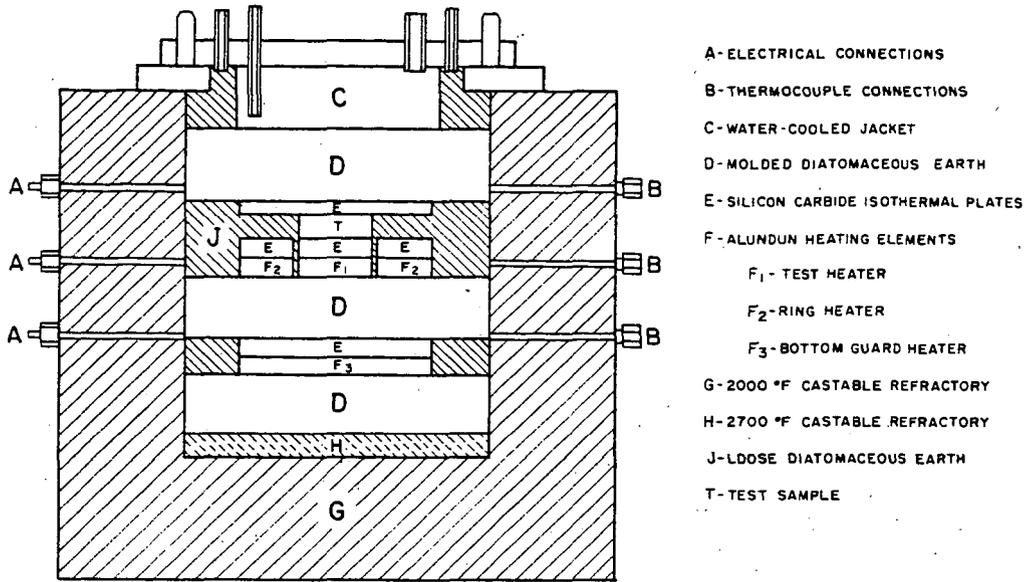
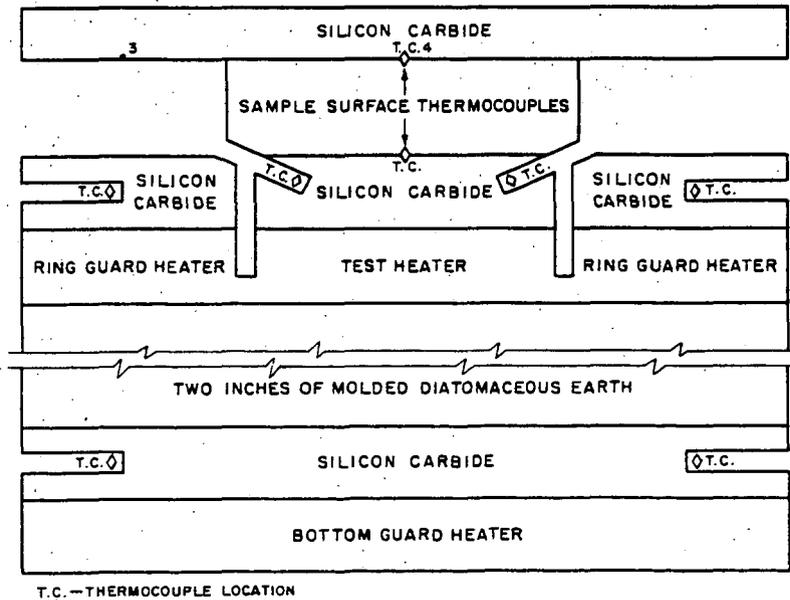


DIAGRAM OF THE THERMAL CONDUCTIVITY CELL

FIGURE I



THERMOCOUPLE ARRANGEMENT IN CONDUCTIVITY CELL

FIGURE II

THERMAL CONDUCTIVITY VS MEAN TEMPERATURE FOR BRIQUETTES MADE FROM ANTHRACITE, BITAMINOUS COAL AND PETROLEUM FLUID COKE.

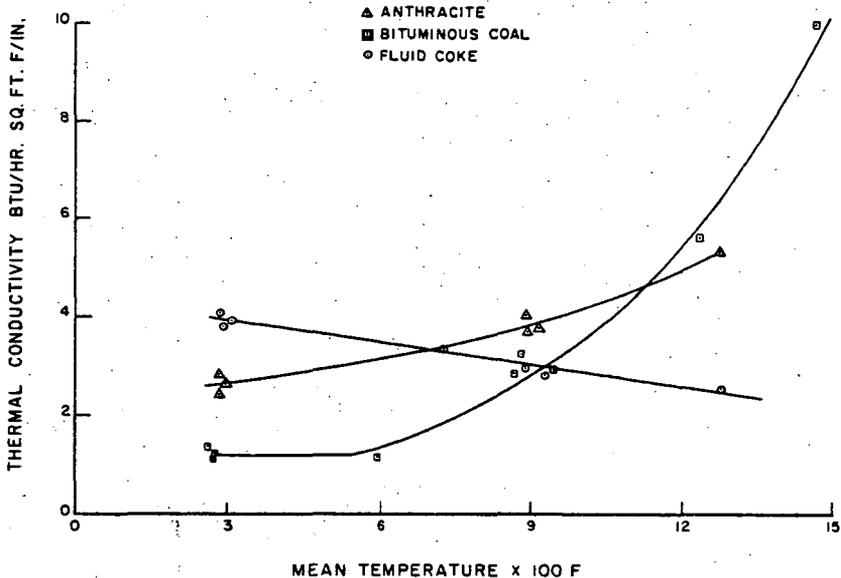


FIGURE III.

COMPARISON OF THE THERMAL CONDUCTIVITIES FOR BITUMINOUS COAL FROM LITERATURE

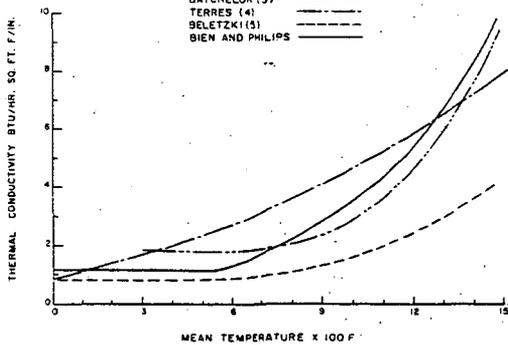


FIGURE IV

COMPARISON OF THERMAL CONDUCTIVITY FOR BITUMINOUS COAL BRIQUETTES BY NORMAL PROCEDURE AND CARBONIZING TO ELEVATED TEMPERATURE AND MEASURING CONDUCTIVITY AT LOWER TEMPERATURE.

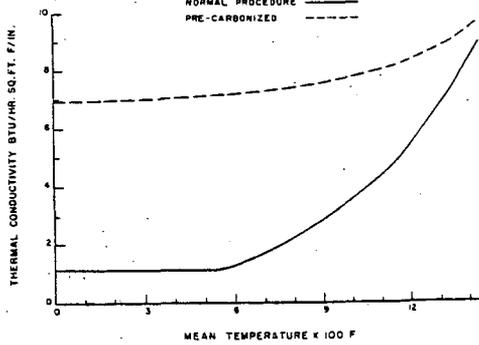


FIGURE V

THE COMPACTION OF SEMI-PLASTIC MATERIALS

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1. INTRODUCTION

For many years coal research scientists have been trying to find an economic method of briquetting bituminous coal without a binder,^{1,2,3,4}. The conventional briquetting processes for coal involve the use of pitch or bitumen as an adhesive; this is undesirable for obvious reasons.

While it has for many years been possible to briquette bituminous coal without a binder, no process has been operated commercially for any length of time, either because the preparation requirements for the coal were too stringent - for example, the coal had to be micronised - or because the compaction pressures demanded were too high for economic operation.

Research at the Coal Research Establishment of the National Coal Board at Cheltenham has sought to remove the need for extremely fine grinding of the particle and the need for the comparatively high pressures of 12 to 15 tons per square inch. A technique has been evolved which works well with British bituminous coals and it has now been applied to many coals from the Commonwealth.

This compaction technique is useful not only with coal: indeed the small financial margins which are available in the coal industries of the world limit this application of the process. After all, where the total process cost cannot exceed 20/- to 30/- per ton, there is no possibility of refined techniques which require machines of some complexity.

It has been shown that the process can be applied to good effect upon catalysts as used in the petroleum and chemical industries, to reactor graphite, iron ore, and many other materials which are loosely termed "semi-plastic".

This paper considers a few aspects of this research and describes some developments which have led to the building of small pilot plant.

2. THE PRINCIPLES OF COMPACTION

In order to obtain a compact of high strength it is necessary to ensure that the compaction technique is effective in bringing about the necessary increase in density and that the compact suffers no damage during extraction from the mould. With coal, at any rate, the strength of a compact for any given size distribution and any given mode of preparation is related to the density of the compact. The density and the strength are determined by the pressure which is applied, but ultimately they approach limiting values which are not exceeded by further increasing the pressure. The limiting density of the compact falls short of the density of the material of the powder by an appreciable margin, say 4 to 20 per cent, depending on the material used (see Fig. 1).

This failure to achieve complete compaction arises in two ways. First, as the briquetting pressure is applied it is opposed by forces set up in the powder and by frictional forces on the walls of the mould which resist the movement of the particles and prevent intimate contact between the surfaces of the particles. Second, when the external pressure is removed the deformed particles recover their shape elastically, in part at least, and the compact expands; the voids within it increase and residual strains remain. A large elastic recovery or a low Young's Modulus is associated with a weak compact.

It has been found that the density of a powder compact can be materially increased for a given applied pressure, if, whilst still under load, it is subjected to shear strain: the strength is increased and the elastic recovery is reduced. The gain in strength with the application of additional shear strain under load may be substantial but the full benefit is obtained only if the shear strain is introduced under maximum load.

The compact made in this way may possess greater density and strength than the limiting values obtained by simple pressing, but whether the compact is made by simple pressing or by introducing additional shear strain under load, the strength and density are still related by the same single curve.

The usefulness of additional shear strain varies with the rheological properties of the material. In the case of a very plastic substance having a negligible elastic recovery, shear strain has little to offer. At the other extreme, anthracite dust or silica sand, both of which are highly elastic and show a very great elastic recovery, say 30 per cent, form no compact at all with or without additional shear strain. The advantage of the process is found to be with materials lying between these extremes, materials which may be termed semi-plastic. Coal is a typical example. The introduction of shear strain improves the briquette by some 5 to 15 per cent as measured by porosity, and the strength is increased by a factor between 3 and 15 (see Figs. 1 and 2).

The meaning of shear strain as applied to an elastic body is well understood (ref. 5, and see Fig. 3a) but its precise meaning when applied to a particulate mass is not so obvious. Consider a mass of coal particles in which spherical lead shot are embedded to form a matrix. When pressure is applied to this mass of particles compaction will occur, and at a specific pressure the lead shot will plastically deform to register the deformation of the particles in the neighbourhood of the shot. The spherical shot will have become ellipsoidal and a measurement of their ellipticity will give the angular shear strain which has occurred above the threshold of pressure at which the shot began to deform.

There are several ways in which the ellipticity can be measured and the angular shear strain thus derived. For example, the compact may be made in an aluminium mould which is X-rayed at various pressure levels; the X-ray photographs will show the ellipticity of the shot from which the shear strain can be calculated, or by a somewhat tedious analysis shear strain may be derived from the translation of the shot. Fig. 3b is a series of X-ray photographs showing the deformation of lead shot in such a briquette.

It is possible in the laboratory to make apparatus which will apply a known amount of shear strain more or less uniformly throughout a mass of particles: such an apparatus is a rotary shear box (see Fig. 4) or an annular shear box (see Fig. 5). Experimental work with this type of apparatus permits a fairly accurate examination of the effect of shear strain to be made and is typical of the apparatus used to obtain the compaction curves previously mentioned. There is a theoretical limit to the amount of shear strain that can be introduced;

at high shear strains and with unfavourable stress distributions slip-plane failure results. The onset of slip-plane failure is determined by an equation familiar in soil mechanics:

$$s = \sigma \tan \phi + C$$

where s is the ultimate shearing stress of the briquetted material,
 σ is the principal stress normal to the plane of failure,
 ϕ is the angle of internal friction,
 C is the cohesion of the material.

The locus of the slip plane is formed by a series of points at which the value of s in the above equation is exceeded by the imposed shear stress. An examination of the photograph of the lead shot markers shown in Fig. 3b will show the onset of slip-plane failure at A. Here the slip plane passes through the shot itself, which has been torn apart by the excessive shear stresses present at that point. This photograph also illustrates the very large elastic rebound which causes difficulty in extraction: unless the compaction forces are released evenly the large differential expansion which results will cause obvious or incipient cracking in the briquette. Additional shear strain improves compaction and reduces elastic rebound, but in order to obtain the greatest benefit the additional shear strain must take place at or near the maximum pressure level.

Shear strain applied at pressures below two-thirds the maximum is, in the case of coal, valueless. Fig. 3c shows the effect of shearing at low pressure. For an unsheared specimen the briquetting sequence was compaction under increasing pressure along the curve a to f, followed by elastic recovery and expansion along the curve f, g, as the briquetting pressure was released. For a specimen subjected to a small shear strain early in the briquetting cycle, the sequence was compaction along the curve a to f, as far as b, followed by a reduction in the porosity b to h, whilst shearing takes place at constant pressure. After shearing the briquetting pressure was again increased but the porosity remained substantially constant until the pressure reached the level required to achieve this porosity in the unsheared briquette.

A possible explanation of this is that a specific briquetting pressure is associated with a given area of contact between the coal particles. If this contact area is increased by the application of shear strain, greater pressure can be carried by the particles before they have to bed down further. From this it is seen that to obtain the greatest improvement in briquette quality (as high density implies high strength) additional shear strain must be applied at as high a pressure as possible. If the briquetting pressure is increased after shearing, the briquette may well "forget" that it has been sheared.

The art of making high-density compacts may therefore be summarized as the introduction of the highest possible shear strain at the maximum pressure, the upper limit of shear strain being set by the development of slip-plane failure. This conclusion applies to a wide variety of materials.

3. THE SEVERAL METHODS OF CARRYING OUT THE PROCESS

Two pieces of apparatus of great use in the laboratory have been illustrated in Figs. 4 and 5, but neither of these can be applied to the briquetting of a cheap raw material such as, for example, coal - either because the frictional resistance inherent in the apparatus is too great, or because of mere mechanical complexity, or because the operational sequence is too difficult. Therefore, although these pieces of apparatus produce the most uniform distribution of angular shear strain, and do this at maximum pressure, they are of doubtful value

to a commercial process, and alternatives which are theoretically not so desirable have to be used.

The germ of a commercial process is to be seen in a two-stage compaction process in which a mass of particles placed in a mould is first compacted with a plane-ended punch and then with a punch profiled as in Fig. 6. The first compact made in the mould exerts a very considerable lateral thrust on the walls of the mould, and therefore although the plane punch is removed, the particles are still under considerable load when the profiled tool is pressed into the first-formed briquette. The shear strain which then occurs does so under very considerable pressure. This method of manufacture was used to form the lead-shot-marker briquette of Fig. 3b. From this photograph it will be seen that the distribution of shear strain is by no means uniform: the centre core undergoes very high shear strain whereas the corners are friable.

This method of compaction can be developed into a commercial machine in which a composite plunger is used to form a powder into a briquette. Fig. 7a shows a simple form of composite plunger. This has two working parts, an outer annulus and an inner core, which are locked together hydraulically until the average pressure over the cross-section of the briquette has risen to a predetermined level near the maximum (4 to 6 tons/sq.in.). At this point oil is allowed to pass from the locking cavity to a reservoir, the core of the plunger moves forward in relation to the annulus and so causes the required angular shear strain. The setting of the pressure level at which oil is exhausted from the locking cavity is important because shear strain must be made to occur under the highest possible loading. Briquettes made by this duplex plunger possess the qualities of hardness expected from them, but there remains the problem of extracting them from the mould without damage, and this, indeed, poses more difficult problems than those associated with their compaction.

It has been noted already² that differential release of load will lead to incipient or obvious cracking of the compact: this, as well as air entrainment, is a frequent cause of "decapping". The only solution is the uniform release of residual strain. An anvil jack is therefore arranged to co-operate with the duplex plunger on its return stroke so that the briquette is subjected to a small longitudinal thrust while it is extracted from the mould. The Poisson ratio effect comes into play and release of strain is very nearly uniform. In the improved plunger shown in Fig. 7b the briquette is made in the extension of the annulus. This arrangement has the advantage that the briquette, when undergoing its initial compression before the application of shear strain, is virtually pressed from both ends so that its density is more uniform and the shear strain, when applied, is more useful. It has the further advantage that extraction is made easier and the anvil jack, essential to the first system, is eliminated. During extraction the centre core of the plunger has to move forward to maintain the longitudinal load upon the briquette at 2 tons \pm 10 per cent while the sides formed by the annulus are withdrawn. This still presents a difficult hydraulic problem.

Each stroke of the duplex ram produces one briquette which may weigh $\frac{1}{2}$ lb. A commercial press has to produce these at many tons per hour. One design of such a press comprises an arrangement of four duplex plungers operating in moulds set in two twelve-station rotary tables. This permits powder to gravity-fill over three stations while it is pressed at another station and ejected at a fifth. The whole assembly is hydraulically operated,

tolerances are small and the speed of operation has necessarily to be high. A cycle time of 1 second was the aim set for a large 5 ton/h experimental version illustrated in Fig. 8. It is not yet a reliable machine but it has produced briquettes of excellent quality at 5 ton/sq.in. pressure.

The hydraulic complexity of the turntable presses based on the duplex plunger made it important to develop simpler methods of applying shear strain under load. It was considered wise to sacrifice both the quality of the product and the low power cost to gain simplicity. The expansion press, shown diagrammatically in Fig. 9a is the result of this policy. The briquette emerges as a long rod which has to be broken into suitable lengths; a simple breaker plate can do this.

The pressure/travel curves of the duplex plunger and the expansion press are compared in Fig. 10. The power cost of expansion is approximately twice that of the duplex plunger; this is accounted for by the high frictional resistance of the briquette rod as it moves through the nozzle.

Apart from considerations of mechanical simplicity, power cost and quality of product, the expansion press has one great advantage over the duplex-plunger press - it is insensitive to fill. The duplex-plunger machine, even though it is hydraulically powered, requires that the variation of fill shall not be greater than ± 3 per cent, whereas the expansion press is equally effective whatever the fill. It is also possible on the expansion press to vacuum-fill; the punch is surrounded by powder which, because the punch is withdrawn at high speed, is forced into the mould under 15 lb/sq.in. pressure and is thereby to some extent compacted. It is surprising that a powder with a free bulk density of 0.4 when fed into a mould in this way has a density within the mould of 0.7.

Because the expansion press is insensitive to fill, and because it is easy to fill the mould, the principle can with comparative ease be applied to a mechanical press (see Fig. 9b) and such a press is probably more suited to the engineering skills available in the briquetting plants of the coal industry than hydraulic presses which need special care in assembly and skill in maintenance. The expansion press is not, however, quite as simple a tool as it appears at first sight. The system is inherently unstable: there is a tendency for the briquetting pressure to drift either up or down so that the machine stalls or briquetting pressure is lost completely. To overcome this, control of the back pressure by a system which is continuous or pulsed is essential. The pressure in the ram applying the lateral force to the four-split flexible nozzle has therefore to be linked via a simple servo control to the pressure developed by the main punch. A typical example of a pulsed control is shown in Fig. 11.

The methods of applying shear strain under load so far described require the use of a reciprocating motion for applying the load, whereas the briquetting of a low-cost raw material is more appropriately carried out in a continuous manner in a double- or ring-roll press. These possibilities have not been overlooked, but it seems unlikely that a pocketed roll can be used to introduce high angular shear strains in this essentially two-stage process. The ring-roll press³, on the other hand, has some promise. The possibility of causing differential slip between the inner roll and outer ring of such a press has been examined but it is difficult to simulate the feed and discharge arrangements without a fairly large-scale press, and this has not yet been done.

4. PROCESS VARIABLES

Before discussing process variables, the influence of which will vary greatly with the material being compacted, it may be wise to consider the mechanism by which high density in a compact may be achieved. Adhesion may well result from a combination of mechanical interlocking, the development of Van der Waal forces, and hotspot fusion or welding. In any event it is necessary to have substantial plastic deformation of the individual particles, in order either (a) to create new clean surfaces and to bring these surfaces sufficiently close together for molecular forces to develop, or (b) to bring about mechanical interlocking.

Under ordinary conditions, many of the particles from which it is required to form compacts, will fail in compression by brittle fracture. If, however, conditions are such that a hydrostatic state of stress can develop these otherwise brittle particles will deform plastically. It is well known, for example, that a carbon rod under tensile test can be made to elongate and 'neck' in the manner of a mild steel specimen if it is subjected to hydrostatic pressure during test.

The technique which has been described of introducing angular shear strain under load favours the plastic deformation of otherwise brittle material by breaking down internal arches within the particulate mass forming the compact and causing thereby the development of a hydrostatic state of stress. But clearly the process variables should be arranged to give the greatest opportunity for plastic flow to develop: moisture content, temperature, particle size, particle shape, will all play their part in this.

Consider moisture content as a single independent variable; in all probability for a porous material the ease with which a particle plastically deforms will increase with increasing moisture content. But at the same time, excessive moisture on the surface of the particle will form a contaminating film to prevent the surfaces approaching one another, and so developing a molecular bond. For this reason there is likely to be, with many materials, an optimum moisture content. Fortuitously, with most coals, this happens to be at or near inherent moisture content level. Moisture will have a secondary effect as a lubricant.

Rising temperature generally increases the ease with which particles deform and unless this rising temperature is associated with, let us say, oxidation of the surface, which will act as a contaminant, increasing temperature will favour the production of a dense compact. Again there is usually an optimum temperature because some adsorption of gas or vapour often occurs to form a contaminant film.

Particle size is of consequence because small particles are shown by microsquashing techniques to deform more readily than large ones. Surface energy considerations may have a bearing upon this. Generally speaking, finely divided material of laminate form will produce the greatest strength and the highest density.

The photomicrograph (Fig. 12a) of a coal briquette shows the plastic deformation which occurs in the large particles of brittle coal giving rise to a laminate structure. There is evidence that the plastic deformation is such that the surfaces of the particles are brought very close together (see Fig. 12b) and you will note the remarkable difference between the structure of the briquette made with and without additional shear strain, (Figs. 12c and 12d).

Fig. 12e is a photomicrograph of a briquette made at an elevated temperature of 420°C at which the exinite has melted or become very plastic indeed to form a matrix holding the more rigid particles together so forming an interlocked structure of great strength.

Increasing pressure will obviously increase the density of the compact to that point at which internal arching and the frictional resistance on the walls of the mould make further increase profitless. It is obviously of economic importance to use the lowest possible pressure and this variable is not therefore of real importance in this discussion.

Restating the obvious: the greatest density and the greatest strength of compact will be obtained by selecting those conditions of moisture content, temperature, particle size, etc., which favour plastic deformation but which do not give rise to the formation of contaminant film upon the surface of the particles.

5. THE COST OF THE PROCESS

The process was developed for use with bituminous British coal and the financial margin was not greater than 20/- to 30/- per ton. It was therefore of paramount importance to devise machines and a preparation technique which were of the least complication. It was essential to look carefully at existing preparation techniques and to develop high speed special purpose briquetting machines. A sample costing for bituminous coal is given in Table I.

Where the financial margins are not so tight, full advantage can be taken of the substantial gains in strength which are available - in the case of coal it was imperative to use simple robust machines which were unable to introduce the shear strain in a uniform manner - process perfection had to be sacrificed to achieve economic simplicity.

6. CONCLUSIONS

Numerous attempts have been made in the past to briquette bituminous coal without a binder, but no process has been commercially successful. In this case research has devised a process which eliminates the need for fine grinding and high pressure previously essential. Machines have been developed for laboratory use and for commercial use in the coal industry of the United Kingdom.

The compaction technique which is described is applicable not only to coal but to semi-plastic materials generally, and it may be that the effectiveness of the technique can be more completely realised with high cost materials such as catalysts and metal powders, than with coal, where only the most crude application of the process can be contemplated.

7. ACKNOWLEDGEMENTS

The work which is described has been carried out at the Coal Research Establishment, Stoke Orchard, Cheltenham, England, under a programme formulated by the National Coal Board, London, and directed by Dr. D.C. Rhys Jones. It is published by kind permission of the Director-General of Research.

The work reported results from the effort of a small research team which has seen the project through from the laboratory to the pilot plant.

The author is indebted to Dr. B.A. Lilley for information in support of Fig. 3c and to Dr. L. Griffiths for his petrographic studies.

Theories of compaction are many and diverse; the opinions put forward, therefore, in this paper do not necessarily reflect the considered view of the Establishment but weigh heavily upon the author's conscience.

8. REFERENCES

1. Sutcliffe, E.R. U.S. Pat. 1,267,711 (1918). Finely ground coal below 200 B.S.S. mesh subjected to high compressive load.
2. Ten Bosch Octrooien N.V. Brit. Pat. 450,633 (1936). A compression matrix for making briquettes or tablets from fine-grained materials or materials in powder form.
Ten Bosch Octrooien N.V. Brit. Pat. 440,811 (1936). Method for the manufacture of briquettes from normally non-coherent substances, particularly coal dust, lignite dust and the like.
3. Spooner, E.C.R. Chem. Engng. Min. Rev., 1949, 403, 41.
Briquetting by Krupp-Herglotz Ring-Roll Press. Fine grinding of coal and pressures of 12 ton/sq.in. - 17 ton/sq.in.
4. Piersol, R.J. Bull. Ill. geol. Surv., 1948, No. 72; Ill. Geol. Surv. Rep. of Invest., 1936, 41. Pt. I, Smokeless Briquettes; Impacted without Binder from Partially Volatilized Illinois Coals.
5. Timoshenko, S. Theory of Elasticity (New York, McGraw-Hill, 1934).

TABLE I

A Sample of Process Costs - United Kingdom - 1959

These costs refer to a commercial plant forming binderless expansion briquettes 2 $\frac{3}{4}$ in. in diameter from high rank coal (volatile matter content 12-20%).

1. <u>Throughput</u>	125,000 tons/year
2. <u>Capital Costs</u>	
Coal Handling	£80,000
Coal drying, grinding, pressing, (including spares)	200,000
Briquette handling	20,000
Instruments	15,000
Site preparation: roads, railway, foundations, buildings, offices, etc.	140,000
10% Contingency	50,000
	<u>£505,000</u>
Interest on capital during construction - 5% for 6 months	£12,000
Working capital, 10% @ 100/- per ton	60,000
	<u>£577,000</u>
Capital investment per ton/year	£4.6/ton/year
3. <u>Operating Costs</u> (per ton of input)	
Wages	2/9d
Power and Heat	4/6d
Repairs and Renewals	1/9d
Overheads and General Expenses	1/6d
Depreciation (plant written off over 10 years)	8/0d
Interest 5%	5/0d
	<u>23/6d</u>

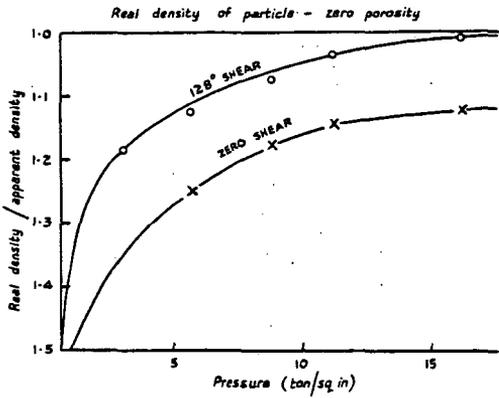


FIG.1. - The relationship between briquetting pressure and density of briquette.

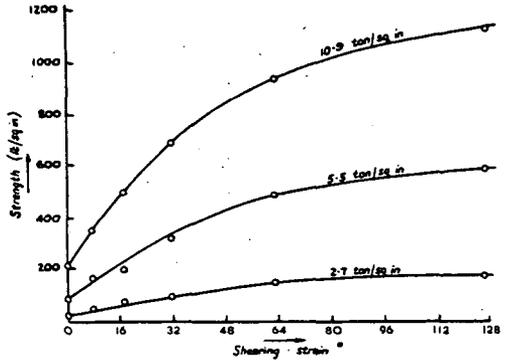


FIG.2. - The variation of briquette strength with applied shear strain.

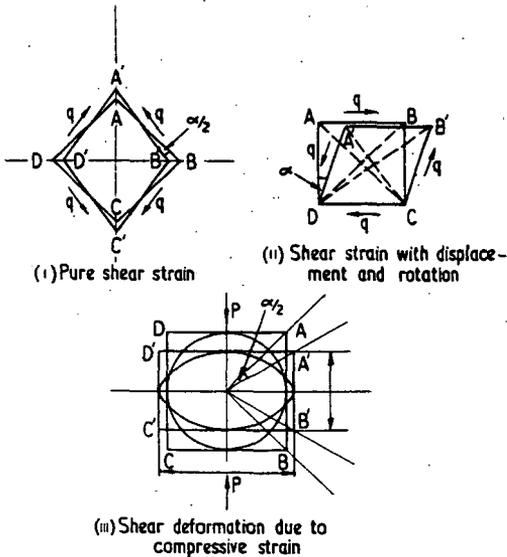


FIG.3(a). - Shear strain - defined as α .

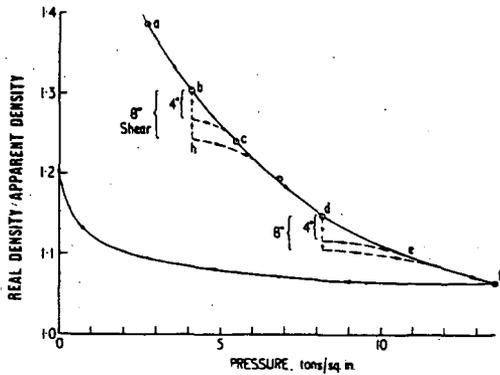
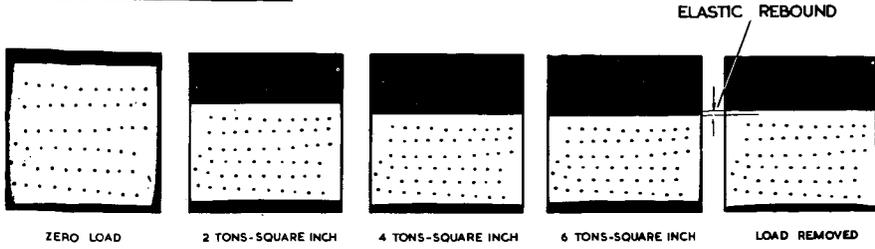


FIG.3(c). - The effect of shearing at low pressure.

① FIRST PRESSING WITH FLAT PLUNGER



② SECOND PRESSING WITH SHAPE PLUNGER

SEE LARGER PHOTOGRAPH

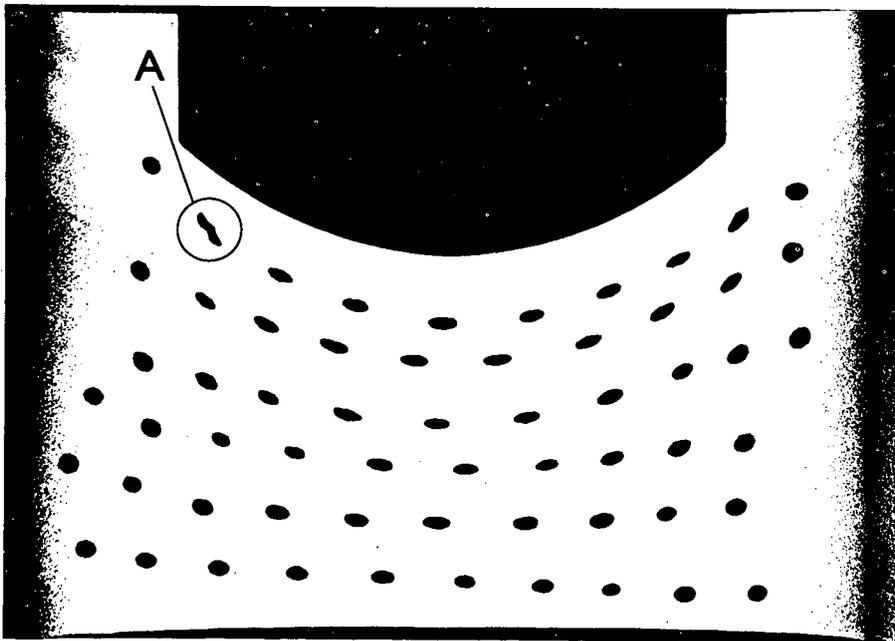
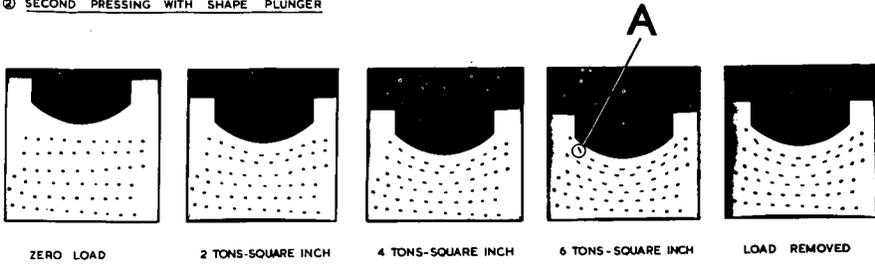


FIG.3(b). - Photograph by X-ray of lead-shot markers in two-plunger briquette.

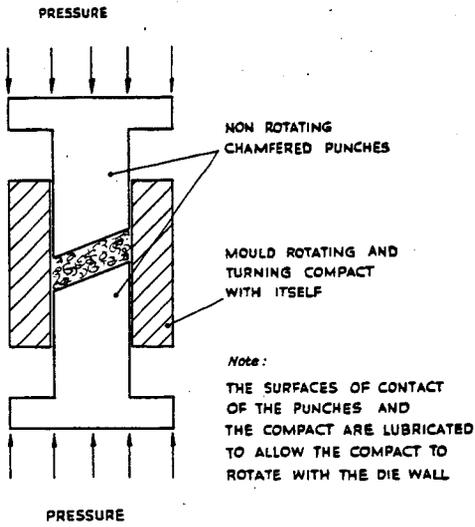


FIG.4. - The rotary shear box.

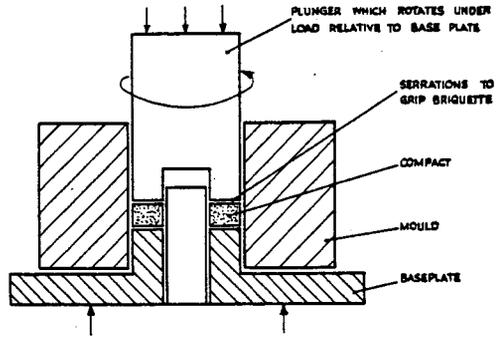


FIG.5. - The annular shear box.

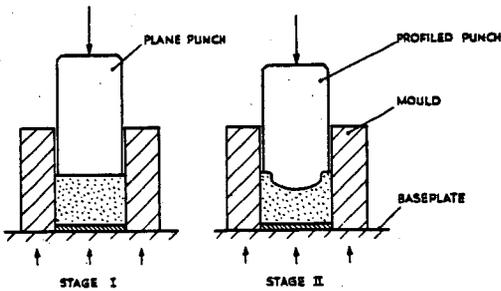


FIG.6. - The double-plunger method.

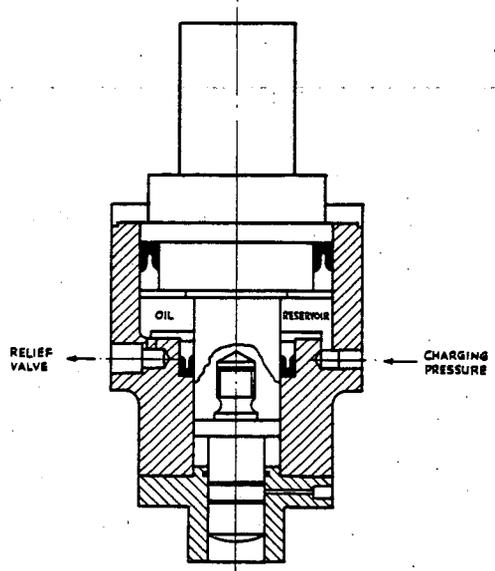


FIG.7(a). - A simple form of composite plunger NK I.

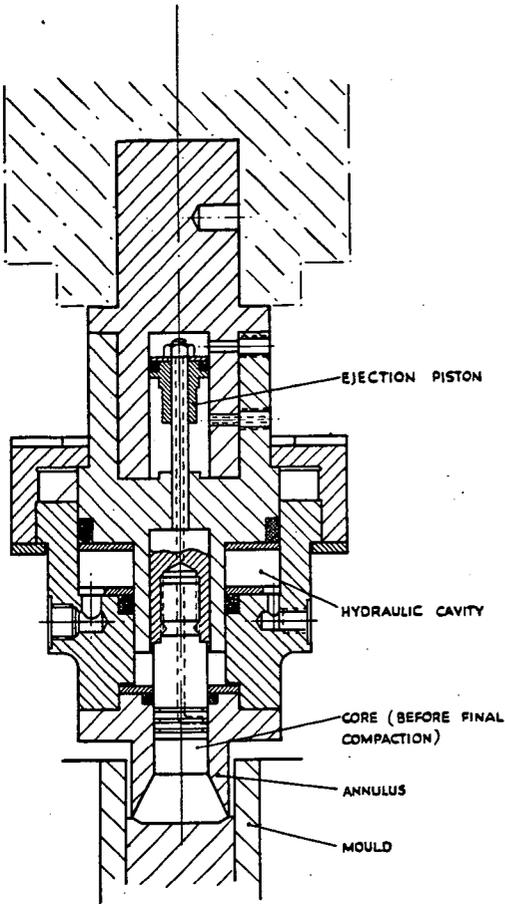


FIG.7(b). - Improved composite or duplex plunger MK II.

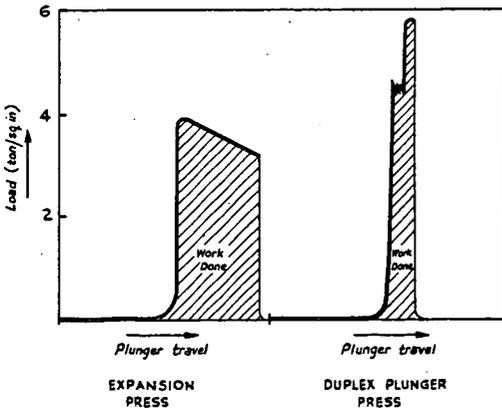


FIG.10. - Pressure-travel curves of duplex plunger and expansion press.

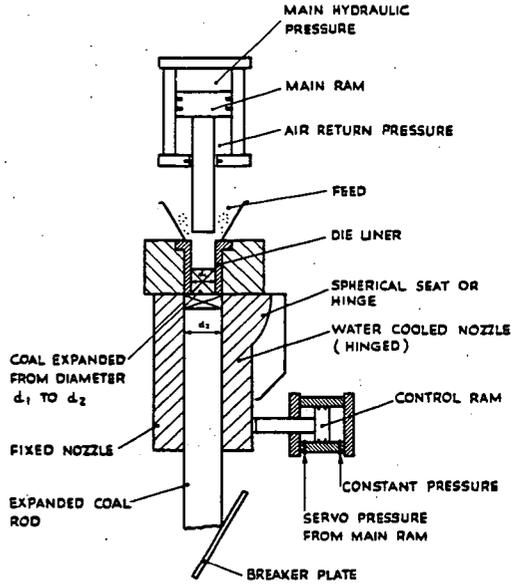


FIG.9(a). - Diagrammatic illustration of expansion principle.

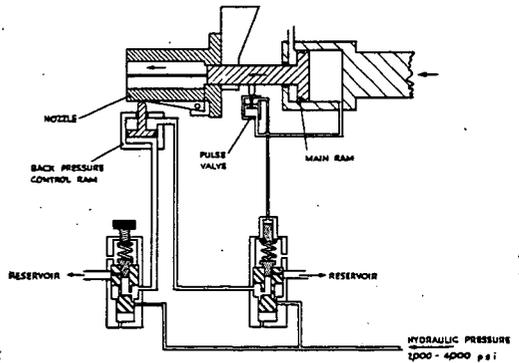


FIG.11. - A back-pressure control system - pulsed control.

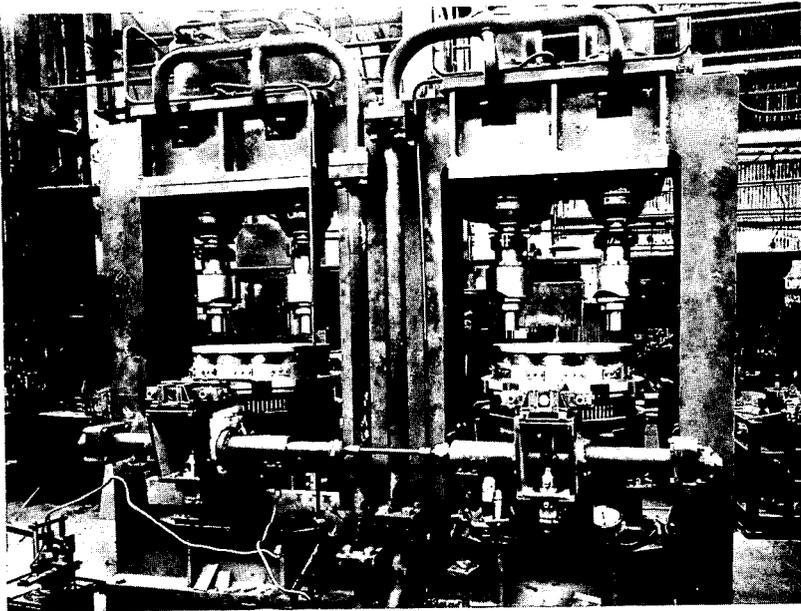


FIG.8 - Hydraulic plunger press.

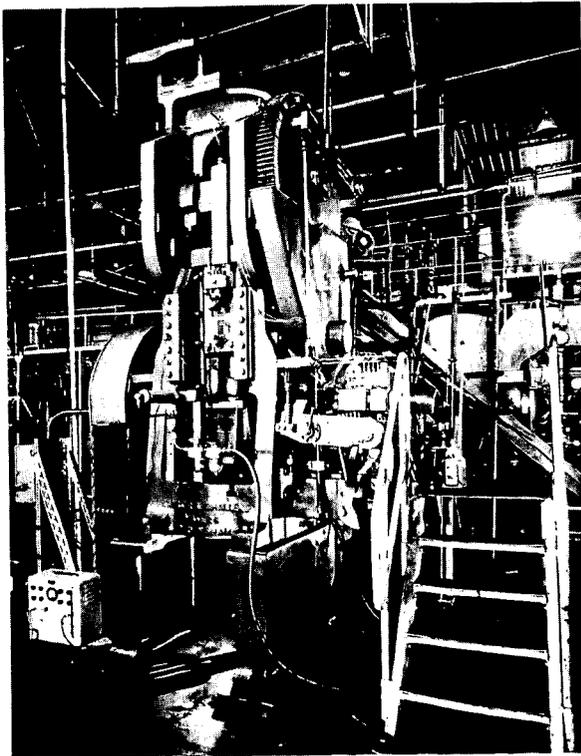


FIG.9(b). - Mechanical expansion press.

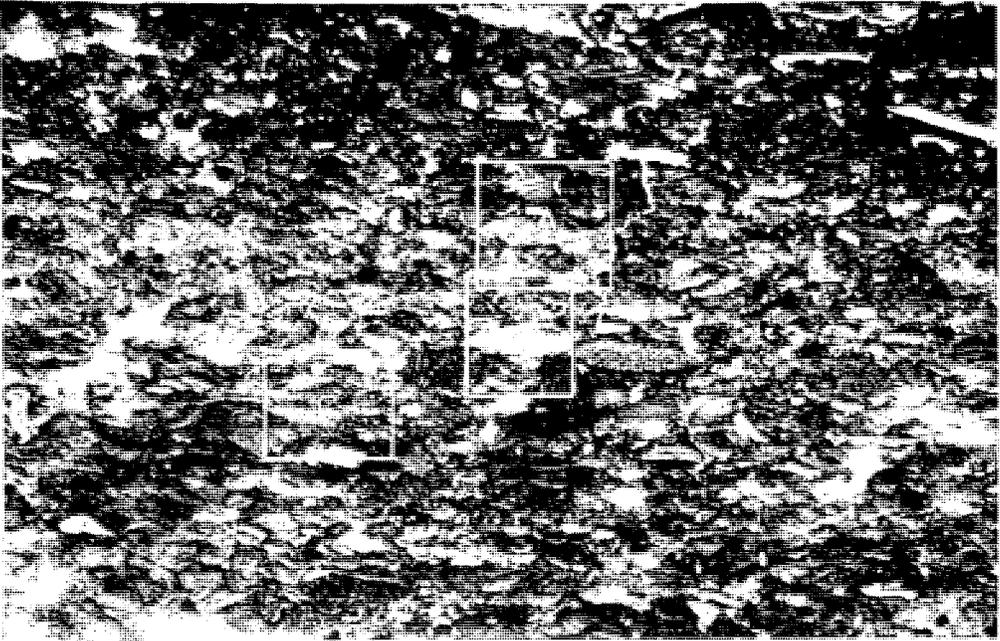


FIG.12(a). - Photomicrograph of Tirpentwys briquette showing deformation of vitrain particles A, B and C and grain. Orientation (dry) magnification 33.

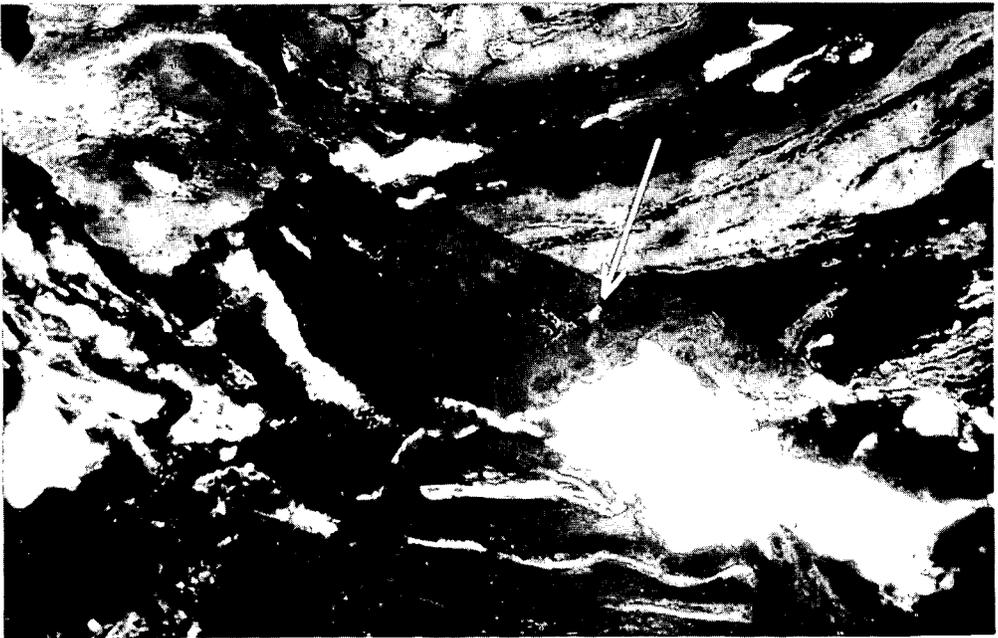


FIG.12(b). - Photomicrograph of Binley briquette made at 110°C showing plastic deformation of vitrain with evidence that the particles are within 0.2 microns (the limit of resolution) (oil, thin section). Magnification 310.

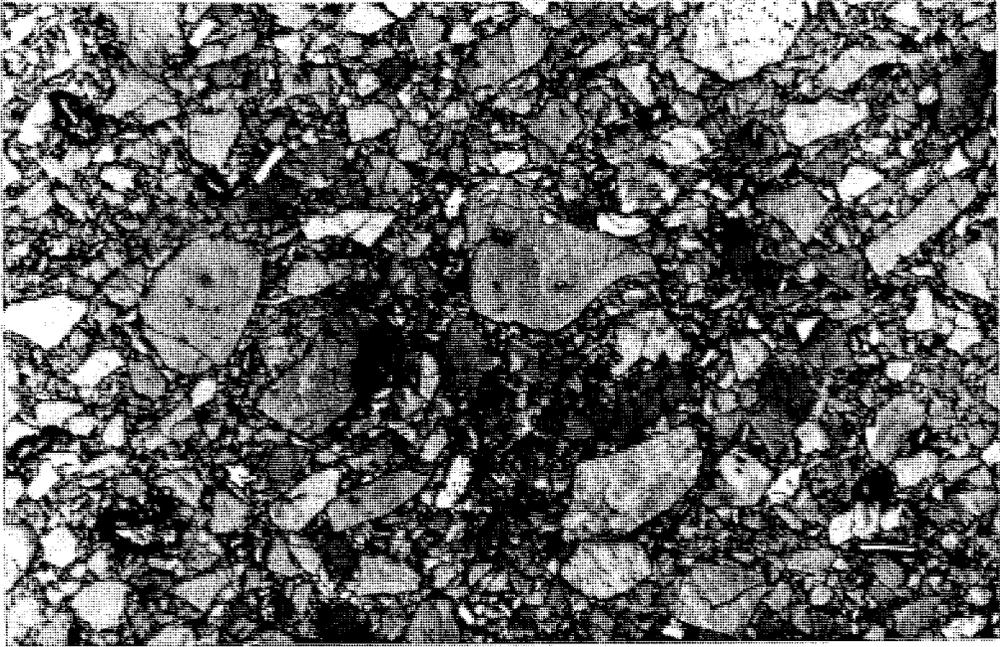


FIG.12(c). - Photomicrograph of coal briquette:
no additional shear strain.

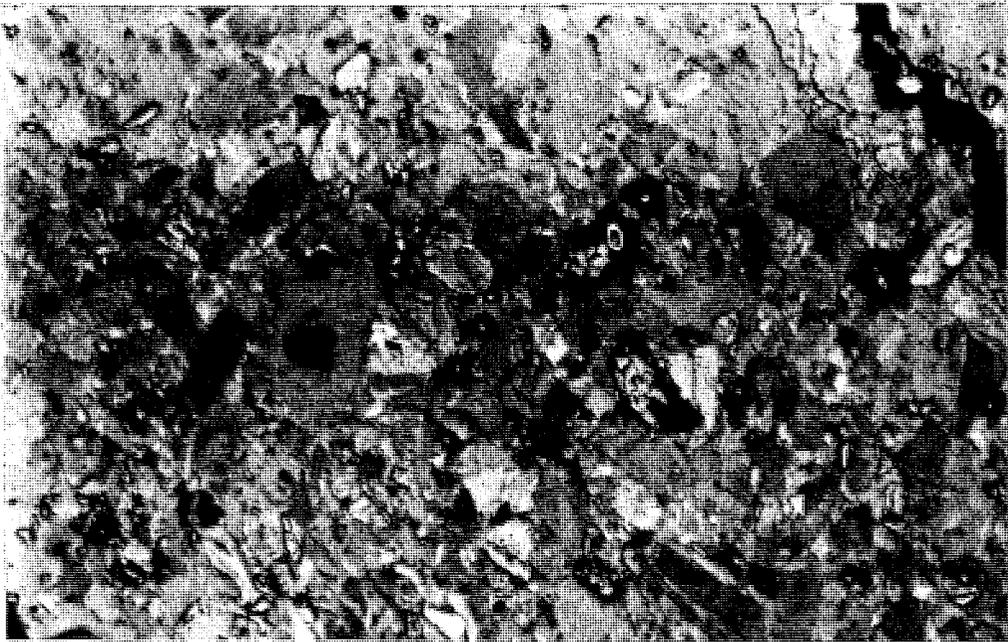


FIG.12(d). - Photomicrograph of coal briquette:
72° of additional shear strain.

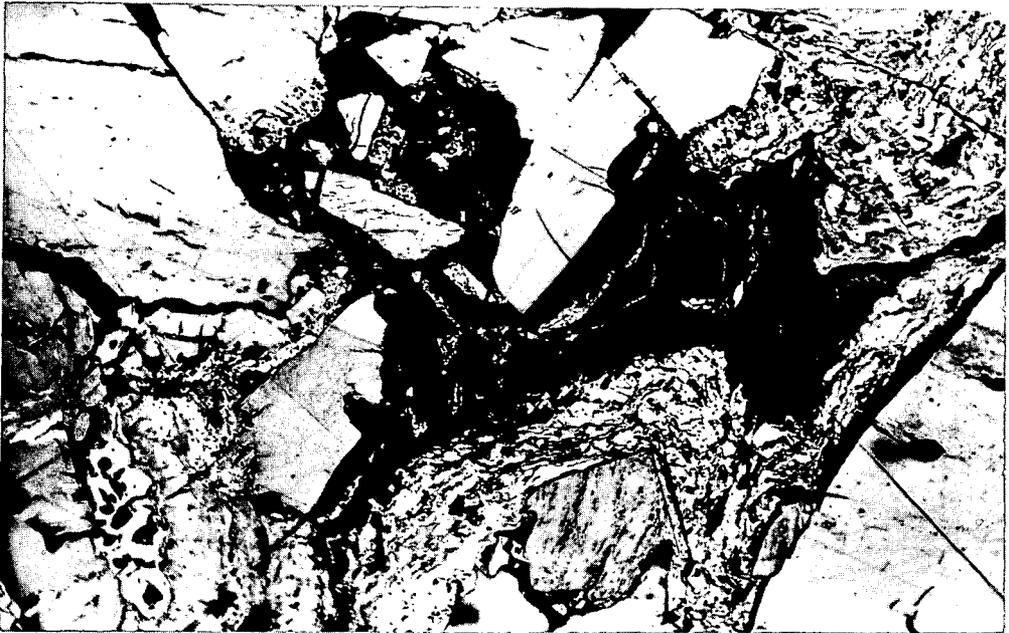


FIG.12(e). - Photomicrograph of briquette made at 420°C showing exinite bridging (oil). Magnification 130.

BASIC NITROGEN IN COALS

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INTRODUCTION

The organic structure of a wide range of coals contains about 1.5 to 2.0 per cent of nitrogen, but little is known about the type of grouping in which this occurs.

Previous investigators found that extraction of a lignite with boiling dilute acid removed about twenty per cent of the nitrogen, mostly as amino acids (1), but only 5 per cent of the nitrogen of higher-rank coals could be identified by this method. Solvent extracts of a bituminous coal were found to be only slightly soluble in aqueous acid (2). Pyridine carboxylic acids are formed at an intermediate stage in the Kjeldahl nitrogen determination (3) and heterocyclic nitrogen compounds are present in distillation products of coals. Hence it has been assumed that much of the nitrogen is present in cyclic structures. However, heterocyclic nitrogen could not be detected by exhaustive methylation of a coal, even after hydrogenation (4).

The sorption of perchloric acid from an anhydrous perchloric-acetic acid mixture (5) has been used in these laboratories for the estimation of basic groups in pitch and tar fractions (6). The same reagent can be used for the detection of basic groups in coals, if the coal is first dissolved or dispersed in an acidic or neutral solvent. Phenol, α -naphthol, and phenanthrene were found to be suitable for this purpose. The perchloric acid reacts with primary, secondary, and tertiary amine groups, including cyclic bases of the pyridine type, but the very weakly basic or acidic nitrogen structures of the thiazole, oxazole, pyrrole, indole, carbazole, or triazole types are not determined.

EXPERIMENTAL

The ultimate analyses of the coals and vitrains examined are given in Table I. The finely ground coal (0.4 gram, minus 200 B.S. mesh) and phenol (10 grams) were gently boiled together under reflux for two hours under nitrogen, then cooled and washed into the titration vessel with 40 ml. of glacial acetic acid. 5 ml. of 0.2 N perchloric acid in glacial acetic acid was added and after 10 minutes the excess perchloric acid was titrated potentiometrically with pyridine (0.2 N) in glacial acetic acid, using a combined glass-calomel electrode. The basic nitrogen content of the coal was calculated from the amount of perchloric acid consumed.

It was established that none of the perchloric acid was consumed by oxidation of the coal: the same titration value was obtained with one coal after the perchloric acid had been replaced by sulphuric acid in acetic acid.

In the determination of total nitrogen the accuracy was ± 1 per cent of the total. A similar level was reached in the titrations, and the values of percentage basic nitrogen recorded in Fig. 1 are thus accurate to within about \pm two per cent for the bituminous coals.

DISCUSSION AND CONCLUSIONS

Fig. 1 shows that for a wide range of coals about fifty to seventy per cent of the nitrogen is in a basic form. When the vitrain samples with carbon content 83.6 to 88.4 per cent were dispersed in phenol, or α -naphthol, 57 to 62 per cent of the nitrogen was found to be basic, the percentage rising regularly with increasing carbon content. For vitrains containing more than 89 per cent carbon the basic nitrogen value appeared to be much lower. An interesting difference was noticed when the same set of vitrains was titrated after being dispersed in boiling phenanthrene. With increase in carbon content from 83.6 per cent to 88.4 per cent the nitrogen content was found to decrease regularly from 32 per cent to 21 per cent, but it then rose to a maximum of 54 per cent in a vitrain containing 89.5 per cent carbon. The basic-nitrogen values for this last sample and for the vitrains of still higher rank were greater when phenanthrene was used instead of phenol as the dispersing agent. This result doubtless reflects the greater solvent power of phenanthrene for high-rank coals. In α -naphthol, high- and lower-rank bituminous coals gave almost the same values of basic nitrogen as in phenol.

The coals containing less than 80 per cent carbon were not petrographically homogeneous and showed a wider scatter of results than the vitrains. Even so, the proportion of the nitrogen in coals that is basic does not vary widely in a range from brown coal to high-rank coking coals.

Sharp end-points were obtained in the back titration of the excess perchloric acid except with the brown coals, for which originally very high values of basic nitrogen had been obtained, some being apparently greater than 100 per cent of the total nitrogen. This effect was thought to be due to the presence in the coal of inorganic ions ("humic" salts), since it is known (7) that metallic salts of carboxylic acids titrate as bases under the conditions of the experiments. The brown coals examined contained about two milli-equivalents per gram of carboxyl groups, partly neutralized as inorganic salts (8). After treatment of the brown and sub-bituminous coals with dilute aqueous acid, sharper end-points were obtained and the observed values of basic nitrogen were 50 to 73 per cent of the total.

The titration was not affected by the presence of carbonate minerals in some of the coals; such salts are not decomposed by the anhydrous acid used.

On an atomic basis, the total nitrogen groups vary from 0.7 per 100 carbon atoms in a brown coal to 1.7 per 100 carbon atoms in a high-rank coking vitrain. Over the same range, the variation in total oxygen groups is from 26 per 100 carbon atoms to 2.9 per 100 carbon atoms. In the low-rank coals nitrogen forms only a minor proportion of the functional groups, but in the high-rank coking coals oxygen- and nitrogen-containing groups are present in similar amounts. The two types of groups may affect the mode of thermal decomposition of the latter coals during carbonization to a similar extent.

The oxygen is more readily driven off from heated coal than is the nitrogen. With a coal containing about 89 per cent carbon, the variation of oxygen and nitrogen content (expressed on an atomic basis) with increasing temperature is as given in Table II.

The greater retention of nitrogen at higher temperatures is perhaps an indication that, unlike oxygen, it is present in cyclic structures in the original coal.

In conclusion, the present investigation has shown for the first time that a major proportion of the nitrogen in a wide range of coals occurs in the form of basic primary, secondary, and tertiary amine groups. These may occur in cyclic structures.

Efforts are now being made in these laboratories to determine the type of nitrogen-containing groups in carbonized coals.

ACKNOWLEDGEMENTS

The coals used and the analyses quoted were obtained in the course of the assessment of the coal resources of Australia which is being carried out under the direction of Mr. H.R. Brown, Chief, Division of Coal Research, C.S.I.R.O., to whom the authors are indebted for his support and for his advice on the preparation of this paper for publication.

REFERENCES

- (1) C.W. Shacklock and T.J. Drakeley, J. Soc. Chem. Ind., **46**, 478 (1927).
- (2) R.S. Asbury, Ind. Eng. Chem., **26**, 1301 (1934).
- (3) A.E. Beet, Fuel, Lond., **19**, 108 (1940).
- (4) L.V. Ramachandran, P.N. Mukherjee, and A. Lahiri, J. Sci. Industr. Res., **18B**, 514 (1959).
- (5) R.T. Moore, P. McCutchan, and D.A. Young, Analyt. Chem., **23**, 1639 (1951).
- (6) E.J. Greenhow and J.W. Smith, Fuel, Lond., **37**, 354 (1958).
- (7) R. Belcher, J. Berger, and T.S. West, J. Chem. Soc., 2882 (1959).
- (8) J.D. Brooks and S. Sternhell, Aust. J. Appl. Sci., **8**, 206 (1957).

TABLE I

ULTIMATE ANALYSIS AND BASIC NITROGEN CONTENTS OF COALS (PER CENT, D.A.F.)

Coal	Carbon	Hydrogen	Nitrogen	Basic N ₂	Basic N ₂ Total N ₂ (%)	Solvent
Dawson Valley	91.4	4.4	1.9	0.09	5	Phenol
				0.61	32	Phenanthrene
Kemmis Creek*	90.3	5.1	1.6	0.13	8	Phenol
				0.58	36	Phenanthrene
Bulli*	89.5	5.3	1.8	0.47	26	Phenanthrene
				0.98	54	Phenanthrene
Bulli*	88.8	5.4	1.5	0.88	58	Phenol
Tongarra*	88.4	5.4	1.7	0.98	57	Phenol
				1.06	62	α-Naphthol
				0.36	21	Phenanthrene
Bulli	87.7	5.0	1.6	0.76	48	Phenol
Tongarra*	87.4	5.4	1.6	0.99	61	Phenol
				0.32	20	Phenanthrene
Wongawilli*	85.3	5.4	1.9	1.12	60	Phenol
				0.55	30	Phenanthrene
Borehole*	84.4	5.7	2.2	1.29	59	Phenol
Borehole*	83.6	5.4	2.1	1.19	57	Phenol
				1.15	55	α-Naphthol
				0.68	32	Phenanthrene
Callide	79.9	4.4	1.3	0.88	69	Phenol
Collie	79.9	4.1	1.5	0.74	50	Phenol
Callide	78.7	4.2	1.2	0.87	74	Phenol
Ravensworth	78.4	4.8	1.9	1.13	59	Phenol
Muja	75.2	4.2	1.1	0.75	59	Phenol
Collie	74.3	5.0	1.3	0.69	52	Phenol
Morwell	70.3	4.9	0.7	0.43	63	Phenol
Yallourn	67.9	4.6	0.6	0.43	70	Phenol

* Vitrain

TABLE II

Coal	N Atoms/100 C Atoms	O Atoms/100 C Atoms
Coal	1.5	3.3
500°C. coke	2.0	2.5
700°C. coke	1.8	1.7
900°C. coke	1.5	0.4

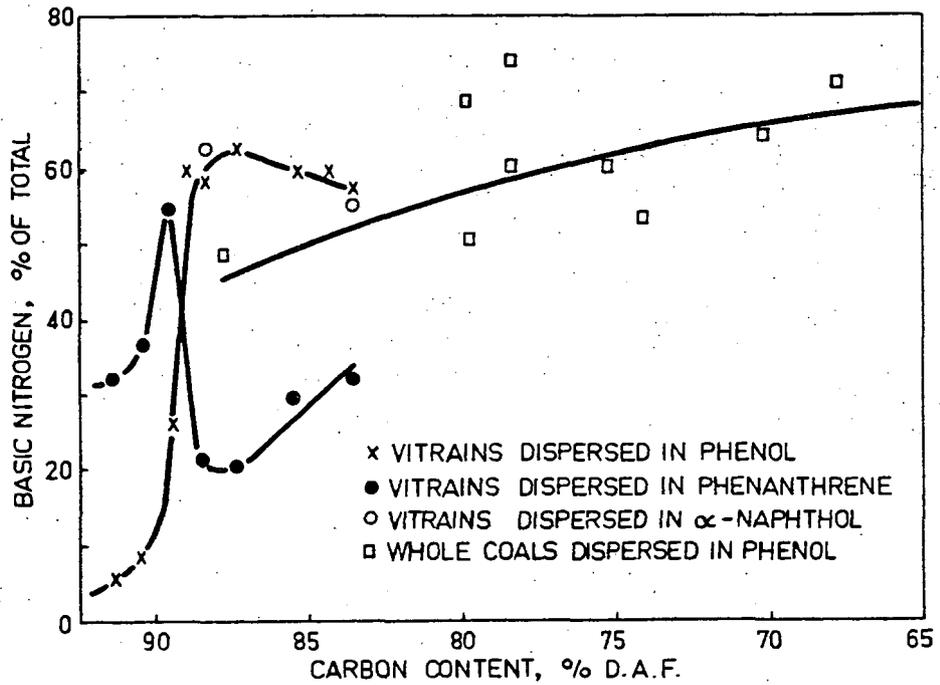


FIG.1.- PROPORTION OF NITROGEN IN BASIC FORM IN COALS

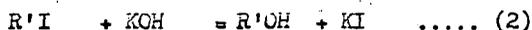
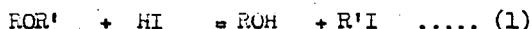
ETHER OXYGEN IN COAL

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Recent work¹ on the state of oxygen in coal structure has established that the major part of oxygen occurs in the form of functional groups, such as -COOH, -OH and >CO. However, a substantial percentage of oxygen, is believed to be present probably as ether or in heterocyclic combination particularly in high rank coals, although direct experimental confirmation is lacking. In the present paper an account is given of the investigations that have been carried out to elucidate the state of oxygen in such combinations.

Treatment of coal with hydriodic acid at an elevated temperature should result in the break-down of ether structure and the ether oxygen will appear as -OH groups. In case of aryl ethers the -OH groups formed on HI treatment will be a direct measure of the ether oxygen. But if mixed ethers are present, the alkali hydrolysis of the HI treated product will result in the formation of further quantity of -OH groups. Therefore the estimation of -OH groups in two stages, as stated above, will give a quantitative measure of the different types of ether oxygen in coal. The course of reaction may be presented as follows :



The above method has been applied to estimate the ether oxygen present in coal. However, it is well-known that in case of unsaturation, the treatment with HI will lead to the addition of hydrogen and iodine to the unsaturated carbon atoms. The iodine thus combined will undergo alkali hydrolysis with the formation of hydroxyl groups. Thus it appears that on the basis of HI treatment and subsequent hydrolysis alone it is not possible to obtain unequivocal evidence regarding the presence of mixed ether in complicated structures like coal, which is likely to contain unsaturation in its structure. To eliminate this uncertainty, unsaturation in coal was estimated by the method developed by the authors earlier².

Experimental

In the present experiments, vitrains from different ranks of coal were selected. The samples were refluxed with hydriodic acid (8 c.c. of HI per gm. of coal, sp. gr. of HI = 1.7) for a period of 8 hours at 130°C and then washed free of hydriodic acid. The product was subsequently analysed for hydroxyl groups.

Estimation of "-OH" groups :- About 1 gm. of the sample was accurately weighed and acetylated with acetic anhydride (20 c.c.) and fused sodium acetate (15 gm.) (by refluxing for 3.5 hours at 140°C. The acetylated product was washed free from acids and hydrolysed by 20 p.c. KOH solution. The product was next acidified with concentrated sulphuric acid and the acetic acid liberated was distilled off and titrated against standard

(see over)

alkali. The p.c. of oxygen as -OH in the HI treated samples was calculated on the initial coal basis.

A second part of the HI treated sample was hydrolysed by aqueous KOH (2.5 N) solution under reflux. The product thus obtained was acidified by hydrochloric acid, washed free of chlorides, and the "-OH" content was subsequently estimated by the above method.

The functional oxygen groups in vitrains were estimated by the conventional methods. The carboxyl and the carbonyl groups in the vitrains were found to remain unaffected by HI-treatment.

For the estimation of unsaturation in coal, oxidation by potash permanganate in dilute aqueous solution was employed. It is known that $KMnO_4$ in dilute aqueous solution brings about hydroxylation of ethylenic double bonds, provided that the reaction is not carried too far and the temperature does not exceed 0°C. The reaction is best carried out in neutral medium. Two hydroxyl groups are formed per one unsaturation linkage.

TABLE - I

Coals	% C	% H	% N	% S	% O (By difference)
A	79.4	5.6	1.9	0.6	12.5
B	83.0	5.8	2.3	0.5	8.4
C	88.0	5.2	2.3	0.5	4.0

TABLE - II

Coals	Total Oxygen %	-OH Oxygen %	-CO- Oxygen %	-COOH Oxygen %	Total reactive Oxygen %	Unaccounted for Oxygen %
A	12.5	6.0	1.6	0.8	8.4	4.1
B	8.4	4.2	0.5	0.2	4.9	3.5
C	4.0	1.2	0.2	0.0	1.4	2.6

TABLE - III

Distribution of ether oxygen in vitrains (expressed on initial d.a.f. coal basis)

Coal	Total unreactive oxygen %	Increase in % oxygen as -OH after HI-treatment	Increase in % oxygen as -OH on subsequent hydrolysis	P.C. Oxygen as ether	P.C. of unreactive oxygen as ether
A	4.1	2.3	2.9	2.3	56.1
B	3.5	2.2	Nil	2.2	62.8
C	2.6	1.6	Nil	1.6	61.5

(see over)

TABLE - IV

Increase in oxygen p.c. as -OH on permanganate (d.a.f. coal basis) oxidation

Coals	Increase in p.c. oxygen as -OH after $KMnO_4$ oxidation
A	2.1
B	Nil
C	Nil

Results and Discussion

The elemental composition as well as the distribution of oxygen in different functional groups of the vitrain samples is presented in Tables I and II. The increase in hydroxyl contents after HI treatment as well as after subsequent alkali - hydrolysis is shown in Table III. It was stated earlier that the increase in OH groups after HI - treatment is to be attributed to the ether linkages, and as such, the increase in oxygen p.c. as "-OH" gives a direct measure of ether oxygen present in coal. In case of mixed ether, however, the alkali hydrolysis of the HI treated product will lead to the formation of additional amount of "-OH" groups.

From an inspection of the Table III it is evident that coals of high rank (B & C) contain only aryl ethers and the increase in oxygen p.c. as "-OH" directly measures the percentage of ether oxygen. In case of low rank coal (A), however, the formation of a significant amount of hydroxyl groups on alkaline hydrolysis probably indicates the presence of mixed ethers. Assuming that the hydrolysis step of the iodo-derivative goes to completion, the increase in -OH contents on alkaline hydrolysis should be equal to that obtained on HI-treatment. The apparent discrepancy between the two values, as evident from Table III, may therefore be ascribed either to experimental errors or to some complicating factor. From Table IV it will be seen that unsaturation occurs in coal of this particular rank. The addition of HI to the double bonds leads to the formation of -OH groups on subsequent hydrolysis. It appears, therefore, that for coals of low rank, the formation of -OH groups on hydrolysis of the HI treated product is partly due to the presence of unsaturation. Thus, for low rank coals, the apparent lack of agreement between the values of hydroxyl groups may be largely explained in terms of unsaturation. It may therefore be concluded that a significant percentage of ether oxygen is present in coals of different rank and that the low rank coals are characterised by the presence of mixed ethers. It would appear that about 60 p.c. of the total unreactive oxygen in coals of different ranks are present in ether combination.

References

1. (a) Innatowichz, A., Komun No. 125, Prace Glownego Instytutu Gornictwa : Katowice, 1952.
- (b) Brooks, J.D., and Maher, T.P., Research, London, 7, 1954, 530.
- (c) Brown, J.K., and Wyss., W.F., Chem. & Ind. 1955, 1118.
- (d) Mazumdar, B.K., Bhangale, P.H., and Lahiri, A., Jour. of Sci. & Industr. Res., 15B, 1, 1956, 44.
2. Mukherjee, P.N., Bhaumik, J.N. & Lahiri, A. - Paper communicated to Chem. & Ind., London.
3. Bhowmik, J.W., Mukherjee, P.N., & Lahiri, A., Fuel, 38, 1959, 211.

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METALLURGICAL COKE MANUFACTURING METHOD
BY BLENDING RAW BRIQUETTE

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1. Introduction

For making metallurgical coke high caking coal is generally blended with coal charge at high ratio.

Many an investigation, 1) have hitherto been made with an intention of saving high caking coal without degrading the quality of coke produced.

The methods attempted in Japan are as follows:-

- (1) Coke breeze blending method. 2)
A small quantity of coke breeze, pulverized under about 0.012" is blended with the coal charge.
- (2) Coalite blending method. 3)
Coalite pulverized under some 0.012" is blended with the coal charge.
- (3) Bojun Tan method. 4)
Low grade coal with high volatile matter is swelled with oil in low temperature.

(1) and (2) were applied to practical operation for some period, but at present they have been stopped using for economical reasons.

There is no prospect of (3) being used in practical operation, notwithstanding it was an excellent investigation. Formerly, we intended to manufacture for many years metallurgical coke directly from low or non-caking coal with high volatile matter, using these coals as main raw material.

At the first stage it was clarified that coking property of these low grade coals was much improved by briquetting under comparatively low pressure (2845-3555lb/in²). Based on this fact, we could produce good metallurgical briquette coke (size: 1.65" x 1.65" x 1.18", strength: stability factor = 40-45 %, hardness factor = 72-74 %) by carbonizing raw briquette at high temperature, when the raw briquette was made from the mixture of low caking coal and non-caking coal reasonably blended as main raw material. The result was already published. 5). In this method the most important point is to keep the quantity of caking constituents of raw briquette within constant limits. It is impossible however to use the ordinary horizontal chamber oven, due to briquette coke (pillowshaped) of the product.

With the object of solving this problem we planned to carbonize the mixture of raw briquette and the coal charge in horizontal chamber oven, by blending raw briquette with the coal charge in coke plant. It is desirable in this case that the coal charge and raw briquette should melt each other during carbonization and good lump coke be produced as the result.

We should like to call this method "Metallurgical coke manufacturing method by blending raw briquette."

The present investigation is in the process of study, and not yet in the stage of discussing its economical value.

Next, mention is going to be made of the main points of this method, as it seems to contain some technically interesting problems.

2. Characteristics of raw coal:

Table 1 shows the characteristics of the representative coals selected out of coals, that are used in a coke plant of Yawata Iron & Steel Works. Pitch in Table 1 is produced in the same plant, and used as binder in making raw briquette.

Table 1: Characteristics of Raw Coals

Items Raw Materials	Proximate Analysis			Fuel Ratio	S (%)	Button No.	Index of Quantity of Caking Constituents*6 (C.I.) (%)
	A.	V.M.	F.C.				
American Coal with medium V.M.	5.02	23.11	71.87	3.11	0.68	8.0	90.5
American Coal with low V.M.	6.70	16.56	76.74	4.64	0.70	6.0	83.6
Kyushu Coal A (low caking coal)	6.89	40.56	52.55	1.30	0.61	3.5	80.1
Kyushu Coal B (low caking coal)	5.64	42.44	51.92	1.22	0.69	5.0	88.9
Pitch	0.50	65.60	33.90	0.52	0.42	-	88.8

In this case, moisture contents of base coal and briquette are severally 8 %, 4 %, and the size of the base coal is <1/4" as in an ordinary case. As seen in Table 2, the proper size of raw coals for briquette is <1/8".

Table 2: Characteristics of Base Coal and Raw Briquette

Items Base Coal + Raw Briquette	Blending Ratio (%)	Screen Analysis of Raw Materials (%)							Proximate Analysis (%)			S (%)	Button No.	Caking Index	
		> 1/8"	1/8" ~ 1/16"	1/16" ~ 0.035"	0.035" ~ 0.023"	0.023" ~ 0.012"	< 0.012"	A	V.M.	F.C.					
Base Coal No.1	American Coal with medium V.M.	40													
	Kyushu Coal A	50	7.5	13.8	10.0	15.0	23.5	30.2	6.83	33.47	59.70	0.64	5.0	88.7	
	Kyushu Coal B	10													
Base Coal No.2	American Coal with medium V.M.	60													
	Kyushu Coal A	30	5.0	12.5	6.5	16.0	21.0	39.0	6.68	30.32	63.00	0.65	6.0	89.0	
	Kyushu Coal B.	10													
Raw Briquette	Kyushu Coal A.	72													
	Kyushu Coal B.	10													
	American Coal with low V.M. Pitch	10 8	0.3	9.9	9.0	10.6	18.6	51.6	6.72	38.22	55.06	0.61	3.5	88.4	

The mixture of high caking coal and low caking coal, prepared so as to contain 40-60 % high caking coal in it is adopted as base coal, with which is blended raw briquette, and the final mixture is the coal charge. Table 2 shows blending ratios and characteristics of two kinds of base coal and raw briquette. And the size of raw briquette is 4/5" (length) x 4/5" (breadth) x 13/25" (height), this has been made specially smaller so that raw briquette may mix well with base coal.

* After 9g of dried coke breeze (48-65 mesh) is mixed with 1g. of dried coal (<65 mesh), the mixture is carbonized at 1742±77.9° for 7 minutes. Coke produced is sorted out by screens of 35 mesh and 48 mesh. Supposing Ag is the quantity of coke over 35 mesh and Bg the quantity of coke, passed 35 mesh but not through 48 mesh screen, the caking index (C.I.) can be obtained by the following formula:

$$C.I. = \frac{A+B}{10} \times 100 (\%)$$

The briquette was manufactured in a pilot plant with capacity of 5 t/day.

3. Size of raw coals for briquette:

Blending ratios of briquette with the base coal (No.1 seen in Table 2) are as shown in Table 3. There are three different sizes of raw coals for briquette: <math> < 1/4'' </math>, <math> < 1/8'' </math>, and <math> < 1/16'' </math>, excepting that size of pitch is always kept <math> < 1/4'' </math>.

After briquette was blended at the rate of 30 % with the base, coke was manufactured in 500 lb. testing coke oven, and then crushing strength and tumbler strength of coke were examined. As shown in table 3, the proper size of raw coals for briquette is <math> < 1/8'' </math>.

Table 3: Relation between Size of Raw Coal for Briquette and Strength of Coke, Blended Raw Briquette

Blending Ratio of Base Coal and Raw Briquette (%)		Size of Raw Coal for Briquette	Strength of Coke (%)		
			Crushing Strength**	Tumbler Strength Stability Factor	Hardness Factor
Base Coal	Raw Briquette				
70	30	<math> < 1/4'' </math>	91.8	53.0	67.8
70	30	<math> < 1/8'' </math>	92.6	52.6	70.3
70	30	<math> < 1/16'' </math>	92.7	53.1	70.4

4. Bulk density of the mixture of base coal and briquette:

Investigating the bulk density on the mixture of base coal and briquette, it was clarified as shown in Chart 1 that the bulk density was developed with the increase of blending ratio of briquette with the base coal, and reached max. value, and then got down.

(Chart 1)

It is expected from the above-mentioned result that the quantity of coal charge per chamber increases by blending briquette with coal charge in practical coke oven.

5. Coke manufacture by a 500 lb. testing coke oven:

Coke manufacture was carried out in a 500 lb. testing coke oven, blending briquette with No.1 and No.2 base coal in various ratios under conditions that one charge is 660 lb., flue temp. of coke oven 2102-2156°F, and carbonization time 17 hrs.

The crushing strength of coke produced is promoted a little, compared with the use of base coal only as shown in Table 4 in case of blending briquette.

Besides, hardness factor increases and reactivity 7) becomes smaller.

From these facts, it is clear that the quality of coke is improved by blending briquette. Moreover, it is favourable that the size of coke has become smaller by this blending.

** JIS K. 2151-1957

Drum ($\phi 59''$ x length 59") rotates in 15 r.p.m. for 2 min., putting 22 lb. of lump coke (>2") in it. Coke is sieved using various screens. Especially 19/32" index is important and the crushing strength is generally indicated by this index.

Generally speaking, the range of crushing strength for metallurgical coke is 91-93 %.

Table 4 : Manufacture of Coke, using 500 lb. Testing Coke Oven.

No.	Blending Ratio of Base Coal. (%)	Blending Ratio of Raw Briquette. (%)	Blending Ratio of Base Coal & Raw Briquette. (%)		Percentage of High Caking Coal in the Mixture of Base Coal and Raw Briquette. (%)	C.I. (%)			Proximate Analysis. (%)				Pushing Strength. (%)			Tumbler Strength. Stability Factor. (%)		*** Reactivity (%)	Average Size of Coke. (int.)
			Base Coal.	Raw Briquette.		A.	V.M.	F.C.	A.	V.M.	F.C.	A.	V.M.	F.C.	Stability Factor.	Hardness Factor.			
			Characteristics of the Mixture (Base Coal + Raw Briquette)			Characteristics of Coke Produced.													
(1)	No. 1		100	0	40	88.7	6.83	33.47	59.70	9.40	0.81	89.79	92.7	57.4	67.6	44.0	2.81		
(2)	American Coal with medium V.M. 40		50	50	25	88.5	6.80	37.27	55.93	9.60	0.72	89.68	92.7	53.1	70.9	33.6	2.80		
(3)	Kyushu Coal B. 10	0	60	40	28	88.5	6.74	36.69	56.52	10.10	0.85	89.05	92.8	51.9	68.8	38.9	2.80		
(4)	Kyushu Coal B. 10	0	70	30	31	88.6	6.44	35.69	57.82	9.60	0.56	89.84	93.3	52.1	70.3	32.6	2.67		
(5)	Kyushu Coal A. 50	0	80	20	34	88.5	6.47	34.71	58.62	9.50	0.71	89.79	93.2	52.2	69.0	38.5	2.68		
(6)		Low V.M.	90	10	37	88.5	6.78	34.76	58.46	9.50	0.81	89.69	93.3	54.7	69.4	40.0	2.68		
(7)			95	5	39	88.6	6.80	34.66	58.54	9.40	0.91	89.69	92.7	55.7	67.5	42.8	2.71		
(8)	No. 2		100	0	60	88.3	6.48	29.96	63.56	8.54	0.58	90.88	93.3	56.8	68.1	38.6	2.80		
(9)	American Coal with medium V.M. 60		90	10	55	88.9	6.60	30.67	62.73	8.50	0.68	90.82	93.3	58.2	68.8	38.0	2.79		
(10)	Kyushu Coal A. 10	Kyushu Coal B. Pitch.	70	30	45	88.8	6.39	32.87	60.74	8.71	0.78	90.51	93.8	56.0	71.2	35.8	2.71		
(11)	Kyushu Coal B. 30	American Coal with low V.M.	50	50	35	88.6	6.25	34.90	58.85	8.92	0.81	90.27	92.6	51.6	70.5	40.2	2.80		
(12)	High Caking Coal. Kyushu Coal 48.5		100	0	51.5	90.1	8.65	31.27	60.08	11.57	1.34	87.09	93.0	55.1	66.1	44.5	3.08		

*** Reactivity is calculated from the composition of CO₂ and CO of gas produced.

As a natural result the blending ratio of high caking coal in the total coal charge has decreased, in proportion to the augmentation of briquette blended. For example, the using rate of high caking coal has been 30 % as shown in the No.(4) in which briquette was blended at a rate of 30 %.

It is recognized from the results that high caking coal is able to be saved to some extent. Moreover, the prototype of briquette was not observed in lump coke on the occasion of blending briquette under 30 %, due to the perfect melting of briquette and base coal during carbonization.

6. Swelling pressure test:

Swelling pressure test was examined for the purpose of comparing the result on the two kinds of samples i.e. mixture, blended briquette with base coal at the rate of 30 % and coal charge (blending ratio: high caking coal 50 %, Kyushu coal 50 %, Yawata Iron & Steel Works in August, 1958).

Swelling pressure was measured in Kopper's movable coke oven 8) under conditions that flue temp. is 2192°F, carbonization time 16 hrs., and one charge 660 lb. Swelling pressure of mixture, blended briquette was a little smaller than in the case of ordinary coal charge, as seen in Chart 2.

(Chart 2)

7. Other tests:

Firstly, quality segregation of coke in a chamber of coke oven was studied on the mixture (blending ratio: base coal 70 %, briquette 30 %), in the case of coke making by blending briquette with base coal. It was explained as the result that there was no particular fear of quality segregation of coke, by mixing base coal and briquette.

Secondly, we tested the by-product on the mixture, in comparison with the case of the base coal only and it was ascertained that yield of tar, light oil in gas and gas has been improved slightly in the case of blending briquette, excepting that the yield of ammonium sulfate decreases.

8. Summary:

Several experiments were carried out to establish the manufacturing process of metallurgical coke, in which while the blending ratio of the high caking coal with the total charge is smaller than in ordinary case, the quality of coke obtained should be not inferior to the ordinary blast furnace coke. For this purpose, the raw briquettes made from low caking coal as main raw coal were blended with the base coal which was almost the same in blending ratio of high caking coal as the coal charge in ordinary coke plant.

The following results were obtained.

- (1) It was ascertained that the proper size of raw coals for briquette to be blended with the base coal was under 1/8". The blending ratios of the base coal and the briquette are as follows:

Base coal (high caking 40 %)	
Medium volatile American coal	40 %
Kyushu coal A	50 %
" B	10 %
Briquette (high caking coal 10 %)	
Kyushu coal A	72 %
" B	10 %
Low volatile American coal	10 %
Pitch (as binder)	8 %

- (2) The bulk density of coal charge (base coal + briquette) increased, by blending the briquette with the base coal.
- (3) The qualities of coke produced in 500 lb. test coke oven by carbonizing the mixture, blended briquettes at the rate of 5-50 % with the base coal, were generally improved by blending briquette in other words the crushing strength and hardness factor were increased and, moreover, the mean size and reactivity decreased. Thus, it is possible for us to foresee that the saving of high caking coal can be carried out by blending briquette with the base coal.
- (4) In the case of blending briquette with the base coal, it was considered that the upper limit of the blending ratio of the briquette was about 30 %, from the observation of lump coke quality and the security of pushing of coke from a chamber in coke oven
- (5) It was attested by the experiment by 500 lb. Kopper's movable wall oven that, when the briquette was blended with the base coal at the rate of 30 %, the expansion pressure during carbonization were in the limits of safety.
- (6) The segregation of coke quality has no trouble in the blending of briquette under above mentioned limits and the yield of by-product is rather favourable by blending briquette with the base coal.

References

- 1) Thionville process
R. Cheradam, *Coke and Gas*, 16 143-9 (1954)
L.F. Minchin, *ibid*, 16 167-171 (1954)
Dry-Charging process
R. Cheradam, *loc. cit.*
M. Perch and C.C. Russell, *Blast Furnace and Steel Plant*, 47, 591-7 (1959)
Stamping process
R. Cheradam, *loc. cit.*
M.S. Patel, *Chem. Eng. Process*, 52, 195-200 (1956)
Sovaco process (L.-B. process)
C. Abramski, *Brennstoff-Chem.* 34, 51 (1953)
E. Burstlein, *Chal. et Industr.*, 14, 354 (1955);
Glückauf, 93 606 (1956)
- 2) Coke breeze blending method
H. Joh, *J. Fuel Soc. Japan*, 26, 98-106 (1947)
S. Tanaka, *Coke Series* (ed. by Fuel Soc. Japan) 1, 51-4, (1950)
- 3) Coalite method
K. Maeda, *ibid.*, 24-32, (1950)
T. Inahara, *ibid.*, 36-42 (1950)
T. Onoda, *Tetsu To Hagane*, 37, 1-7 (1951)
- 4) Bojun Tan Method
T. Saheki and S. Matsuzawa, *J. Fuel Soc. Japan*, 28, 232-250 (1949)
Y. Yoshida et al, *Coke Series* (ed. by Fuel Soc. Japan), 4, 200-212 (1953)
- 5) H. Joh and S. Ida, *Tetsu To Hagane*, 42, 226-8, 910-2 (1956); 43, 1024-6 (1957); *Jap. Pat. No.223776*.
- 6) H. Joh and S. Ida, *Coke Series* (ed. by Fuel Soc. Japan), 4, 28-9 (1953)
- 7) H. Joh and S. Ida, *ibid.*, 60-80 (1953)
- 8) H.H. Lowry, "*Chemistry of Coal Utilization*", 1, 274-5 (1947)
H.W. Jackmann, *Illinois State Geological Survey, Circular 246*, (1958)

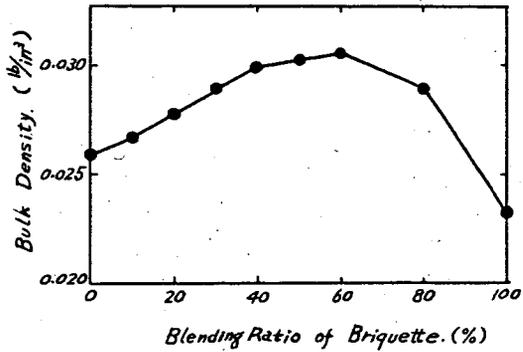


Chart 1 : Relation of Bulk Density and Blending Ratio of Briquette.

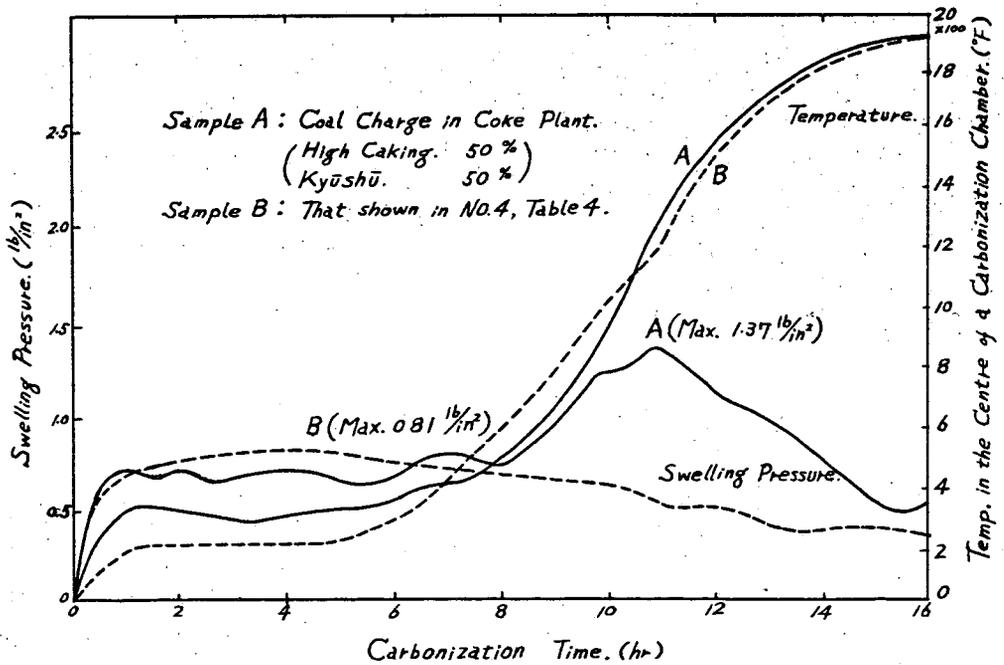


Chart 2 : Comparison of Swelling Pressure.

PILOT PLANT INVESTIGATION ON THE BRIQUETTING AND CARBONISATION OF SOUTH ARCOT LIGNITE

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1. INTRODUCTION

The Neyveli Lignite Corporation Ltd. are now implementing an Integrated Project for the development of lignite resources at Neyveli, Madras State, India. The lignite bearing area here is 100 square miles and the reserves are estimated at 2,000 million tons (1). Tests done at Neyveli and other laboratories have indicated that the lignite is of very good quality and that it could be briquetted without a binder. Table 1 gives the analysis of South Arcot lignite.

In the first stage of development, the Integrated Lignite Project consists of the following schemes:

- 1) An Open Cast Mine, for producing 3.5 million tons of lignite per annum.
- 2) A Thermal Station with an installed capacity of 250 M.W. The Thermal Station will burn pulverised raw lignite as fuel.
- 3) A Fertilizer Plant for producing 152,000 tons of urea per annum by the total recycle process. This plant is expected to be the biggest of its kind in the world.
- 4) A Briquetting and Carbonisation Plant for producing 380,000 tons of carbonised briquettes per annum. This will be one of the biggest lignite briquetting and carbonisation plants in the world. The carbonised lignite briquettes will be used mainly as domestic fuel in South India.

When the Corporation started the development of lignite resources, it did not have adequate data to decide upon the type of equipment that should be used for drying, briquetting and carbonisation of lignite. It was considered risky to base the design of a large commercial unit on laboratory data alone. Too, experiments conducted in Germany showed that it was not possible to conduct pilot plant tests on drying, briquetting and carbonisation at any one place, because all the equipment were not available in any one laboratory. The technical opinion was that the weather had a significant role to play in these processes and the best thing would be to conduct pilot plant tests at Neyveli itself. Accordingly, a pilot briquetting and carbonisation plant was installed at Neyveli. This was erected in a record time of eight weeks and was put into commission on the 14th May, 1958.

2. DESCRIPTION OF THE PILOT PLANT:

The pilot plant consists of a raw coal preparation section, a Parry dryer in which the lignite is dried in an entrained state by hot inert gases, an extrusion type briquetting press made by Messrs. Pawert AG. of Switzerland and a carboniser designed by the U.S. Bureau of Mines. It also has tar and gas collecting systems. The arrangement of the major equipment in the Pilot Plant is shown in figure 1. The plant was supplied by the Technical Co-operation Mission (U.S.A.).

The raw coal preparation section has a hammer mill with a throughput of 3 to 5 tons of raw lignite per hour. The dryer can handle upto 2.5 tons of raw lignite per hour, when drying lignite from 56% to 10% moisture. The briquetting press can make briquettes of two nominal sizes, 65 mm. (2½") diameter and 38 mm. (1½") diameter. The capacity of the press, when it makes 65 mm. diameter briquettes, is about 1 ton of briquettes per hour. When it makes 38 mm. diameter briquettes, the press capacity is of the order of 1000 lbs. per hour.

The carboniser was designed to carbonise continuously about 300 to 500 lbs. of raw briquettes per hour. However, it was found during actual practice that it was difficult to obtain satisfactory performance, when the carboniser is operated on a continuous basis. Such an operation gave too many fines in the product. Batch tests have been done, using charges of the order of 1000 to 2000 lbs. of raw briquettes at a time.

3. DESCRIPTIONS OF THE RUNS MADE IN THE PILOT PLANT

Since the pilot plant came into operation, 26 runs on the dryer, 77 runs on the briquette press and 44 runs on the carboniser have been made. The aim of the investigations was to find out the optimum drying, briquetting and carbonisation conditions for producing good carbonised briquettes for use as domestic fuel in South India. The carbonised briquettes are expected to take the place of charcoal, which is a commonly used domestic fuel in South India at present. Due to scarcity of charcoal, the need for a substitute fuel is keenly felt. Under the prevailing conditions, the fuel that is to replace charcoal has to be, as much as possible, similar to charcoal in handling and combustion properties. Otherwise, there is a likelihood of consumer resistance. The volatiles in the carbonised briquettes have to be low enough to cause no smoke in actual use, but high enough to facilitate easy ignition of the fuel. The carbonised briquettes also must have fairly strong structure, such that they can be transported over a distance of 200 to 300 miles, without appreciable amount of degradation in size.

The variables that were investigated, were the following:-

- 1) Size of the dried lignite for briquetting.
- 2) Moisture in the raw briquettes.
- 3) Size of the raw briquettes.
- 4) Temperature of carbonisation.
- 5) Heating rate during carbonisation.
- 6) Method of quenching of carbonised briquettes.
- 7) Influence of ash on the strength of carbonised briquettes.

Laboratory scale experiments (2,3) indicated that best briquettes for carbonisation could be produced by taking fine dried lignite with as low a moisture content as possible and pressing it with as high a pressure as possible. No binder is required for briquetting. However, some evidence showed that better briquettes were obtained when the dried lignite had a size of 0-2 m.m. rather than 0-1 m.m. (4). The arrangements in the coal preparation section of the pilot plant was such that much control could not be exercised on the sizing of raw crushed lignite. The only control that could be had was on the shape, size and number of hammers in the hammer mill. Initially the mill was designed to operate at 3000 RPM., which gave too fine a material for drying and briquetting. The very fine dried lignite powder that was produced could not be briquetted in the extrusion press. The speed of the hammers was then changed to 1450 RPM., which gave better results. Table 2 gives typical sizes of the product from the hammer mill before and after the changes were incorporated in the mill. The best briquetting results were obtained when the -200 mesh material of crushed raw lignite was of the order of 3 to 4%. When the -200 mesh material was of the order of 9 to 10% in the crushed raw lignite, the -200 mesh material in the product was of the order of 33 to 34%. This dried lignite was too fine to be briquetted in the extrusion press of the Pilot Plant. When the

-200 mesh material in the dried lignite was of the order of 18%, as was the case after the modifications were carried out in the hammer mills, the dried material could be briquetted easily in the press.

Moisture in the raw briquettes plays a significant role in the production of good carbonised briquettes. Laboratory tests indicated that the lower the moisture content in the raw briquettes, the stronger and better were the carbonised briquettes. However, in actual practice there is a limitation to the minimum moisture content in the dried lignite, which is used for briquetting. Very low moisture in the lignite will make it difficult to handle in the conveyors and briquette presses. It also produces a great fire hazard. High moisture content in the raw briquettes will result in weak carbonised briquettes. Sometimes the raw briquettes themselves will be weak. Experiments in the extrusion press installed at Neyveli indicated that when the moisture content in the dried lignite is of the order of 10 to 14 percent, good raw briquettes could be made. Actual experiments in the carboniser have indicated that briquettes with 12% moisture and below give good carbonised briquettes. Table 3 gives the yield of whole carbonised briquettes, when the moisture content varied from 10 to 17%. The data are clear that when the moisture content in the raw briquettes is between 11.7 and 12.7%, the number of whole carbonised briquettes is large.

The size of raw briquettes plays a role in the production of lumpy carbonised briquettes. For actual household use in South India, the briquettes need not be of very large size at all. The preferable thing would be to have a briquette which would be of 25 to 38 mm. in diameter and 25 to 38 mm. in thickness. Two sizes of briquettes were investigated in the Pilot Plant, viz. 65 mm. and 38 mm. (diameter). The thickness of the briquettes in both cases varied from 25 to 40 mm. It was found that invariably the yield of whole carbonised briquettes was higher with the 38 mm. raw briquettes. It was possible, in batch tests, to obtain almost hundred percent unbroken carbonised briquettes with a charge of raw briquettes of 38 mm. diameter. Tables 3 and 4 give these results.

The design of the carboniser was such that it was difficult to measure the exact carbonisation temperature. The measurement was made of what is called "gas cross-over temperature". This temperature varied from 600° F to 1210° F during the tests under discussion. The carbonisation temperature was approximated from the volatile matter content in the carbonised briquettes. When the carbonisation temperature was about 650° C, the volatile matter in the carbonised briquettes was of the order of 15 to 18% (moisture and ash free). These briquettes gave a smokeless flame and had good combustion properties. Lower temperatures yielded carbonised briquettes which had higher percentage of volatiles, which resulted in these briquettes giving smoky flame. It was not possible to raise the gas cross-over temperature of the carboniser beyond 1210° F. This corresponds to a carbonisation temperature of about 750° C. The minimum volatile matter in the carbonised briquettes obtained during these tests was 8.5% (m.a.f.). The rate of heating was critical in all cases. The heating had to be as slow as possible upto about 250° C. Beyond that, the charge could be heated at a faster rate. Figure 2 gives typical heating curves during these carboniser runs. The heating time in these runs varied from 6 to 21 hours.

In commercial operations, two types of quenching are used for cooling the carbonised briquettes, dry quenching with inert gases and wet quenching with water sprays. Dry quenching methods are generally more expensive. Table 5 lists the results obtained during dry quenching and wet quenching of carbonised briquettes made under identical conditions. The investigations showed that as far as the size of the carbonised briquettes is concerned, the method of quenching had no significant effect. The only difference was that wet quenching increased the moisture content

in the carbonised briquettes.

The ash content in the raw lignite has a great influence on the quality of briquettes. The ash content in the raw lignite that was used in these tests varied from 2.8% to 7.3%. As far as can be seen from the data collected during these runs, although the variation in ash content did not have a pronounced effect on the shatter index of carbonised briquettes, yet it greatly influenced the tumbler index. Table 6 indicates these results.

Table 7 gives the physical properties of raw briquettes and carbonised briquettes. The chemical and physical properties of these carbonised briquettes are similar to those produced in Germany (5) and correspond to the properties of charcoal used in South India (6).

4. SUMMARY

Pilot Plant investigation on the drying, briquetting and carbonisation of South Arcot lignite showed that good carbonised briquettes could be produced for use as domestic fuel. Optimum conditions for drying, briquetting and carbonisation were established.

5. ACKNOWLEDGMENT

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6. REFERENCES

- (1) Ratnam, C.V.S., "South Arcot lignite - Part I, Nature", Madras Information, 44, April 1952.
- (2) Ratnam, C.V.S. & Veeraraghavan, "Carbonisation of South Arcot Lignite", J. Sc. & Ind. Res., 13B, 210 (1954).
- (3) Ratnam, C.V.S. & Dorairaj, B., "Studies on South Arcot Lignite Briquettes", J. Indian Chem. Soc., Ind. & News Edition, 19, No. 2, 57 (1956).
- (4) Rammler, E., Bergakademie, Freiberg, Private communication.
- (5) V.E.B. Kombinat, "Otto Grotewahl", Boehlen, Private communication.
- (6) Ratnam, C.V.S. & Ramanathan V.S., "Utilisation of South Arcot Lignite Char as Domestic Fuel", Current Science, 25, 182 (1956).

TABLE 1 - ANALYSIS OF SOUTH ARCOT LIGNITE

a) Proximate analysis:

Moisture	%	51.00 - 56.00
Ash	%	2.20 - 3.30
Volatile matter	%	22.40 - 24.93
Fixed Carbon	%	19.10 - 20.59

b) Ultimate analysis:

Carbon	%	29.48 - 33.60
Hydrogen	%	2.07 - 2.40
Nitrogen	%	0.24 - 0.29
Oxygen	%	8.63 - 9.21
Sulphur	%	0.29 - 0.48
Ash	%	2.20 - 3.30
Moisture	%	51.00 - 56.00

c) Calorific value:

Gross:	k.cal/kg	2560 - 3165
Net:	k.cal/kg	2307 - 2902

TABLE 2 - SIZE ANALYSIS OF CRUSHED RAW LIGNITE AND CORRESPONDING SIZE ANALYSIS OF DRIED LIGNITE

Sieve No. (Tyler)	Size of opening mm.	Before changing hammers and reducing motor speed				After changing hammers and reducing motor speed			
		Crushed raw lignite		Dried lignite		Crushed raw lignite		Dried lignite	
		Sample I	Sample II	Sample I	Sample II	Sample I	Sample II	Sample I	Sample II
		Cumulative weight %	Cumulative weight %	Cumulative weight %	Cumulative weight %	Cumulative weight %	Cumulative weight %	Cumulative weight %	Cumulative weight %
4	4.699	13.4	--	--	--	10.21	--	--	--
8	2.362	28.8	--	--	--	31.05	--	--	--
16	0.991	57.8	3.20	7.45	57.03	6.12	6.63	6.63	6.63
28	0.589	63.2	15.82	22.00	68.37	22.48	24.34	24.34	24.34
48	0.295	76.6	37.22	38.50	79.38	48.38	49.28	49.28	49.28
60	0.246	78.6	42.04	42.15	82.40	52.50	53.19	53.19	53.19
80	0.175	83.0	49.09	48.25	87.25	62.82	63.30	63.30	63.30
100	0.147	86.1	54.41	53.40	93.23	68.44	69.44	69.44	69.44
200	0.074	91.0	66.06	65.82	96.55	81.25	81.03	81.03	81.03
-200	0.074	9.0	33.94	34.18	3.45	18.75	18.97	18.97	18.97
Moisture %		50.3	11.70	9.80	49.2	12.8	12.00	12.00	12.00

TABLE 3 - INFLUENCE OF MOISTURE ON THE YIELD OF CARBONISED BRIQUETTES.
(Size of raw briquettes - 65 mm.)

<u>Moisture in the raw briquettes</u> %	<u>Yield of full briquettes</u> %
17.2	9.7
16.2	17.2
13.6	29.9
12.7	49.6
12.0	53.0
11.7	48.0
11.4	38.0
11.0	35.7

TABLE 4 - YIELD OF WHOLE CARBONISED BRIQUETTES
(from 38 mm. raw briquettes)

Carboniser run	7	8	15	16	17*
Temperature - Gas cross-over °F	775	700	775	700	630
Size of raw briquettes - diameter in mm.	38	38	38	38	38
Moisture in the raw briquettes %	11.4	11.4	11.6	10.4	12.0
<u>Yield of carbonised briquettes:</u>					
+ 25 mm. %	100	100	100	100	94
- 25 mm. %	—	—	—	—	6

*The temperature at the time of charging briquettes in run No. 17 was 460° F. This explains the lower yield of full briquettes.

TABLE 5 - INFLUENCE OF METHOD OF QUENCHING ON THE SIZE AND STRENGTH OF CARBONISED BRIQUETTES.

Run No.	33		34		35		36	
	Gas quenched	Water quenched						
<u>Raw briquettes:</u>								
Moisture %	11.4		11.0		13.2		13.2	
Ash %	14.4		13.8		8.2		8.2	
<u>Carbonised briquettes:</u>								
Temperature of gas cross-over °F	700		700		645		700	
+ ½" % of product	80.2	86.7	80.9	80.7	96.6	96.4	94.1	92.3
Compression strength kg/cm ²	120	112	152	186	181	164	160	167
Moisture in carbonised briquettes %	2.1	4.5	1.5	3.4	1.9	9.0	1.3	4.0

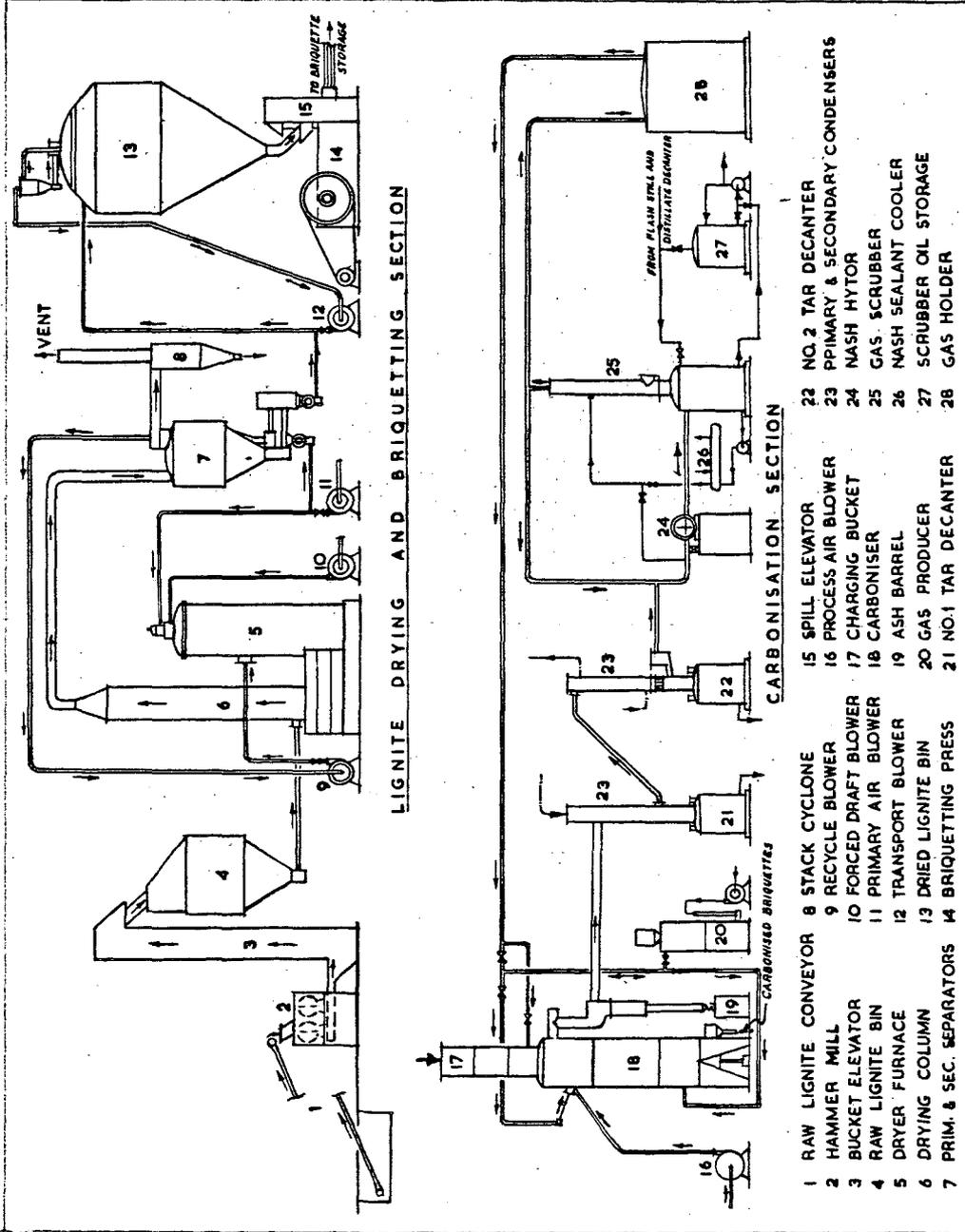
TABLE 6 - INFLUENCE OF ASH ON THE STRENGTH OF CARBONISED BRIQUETTES.

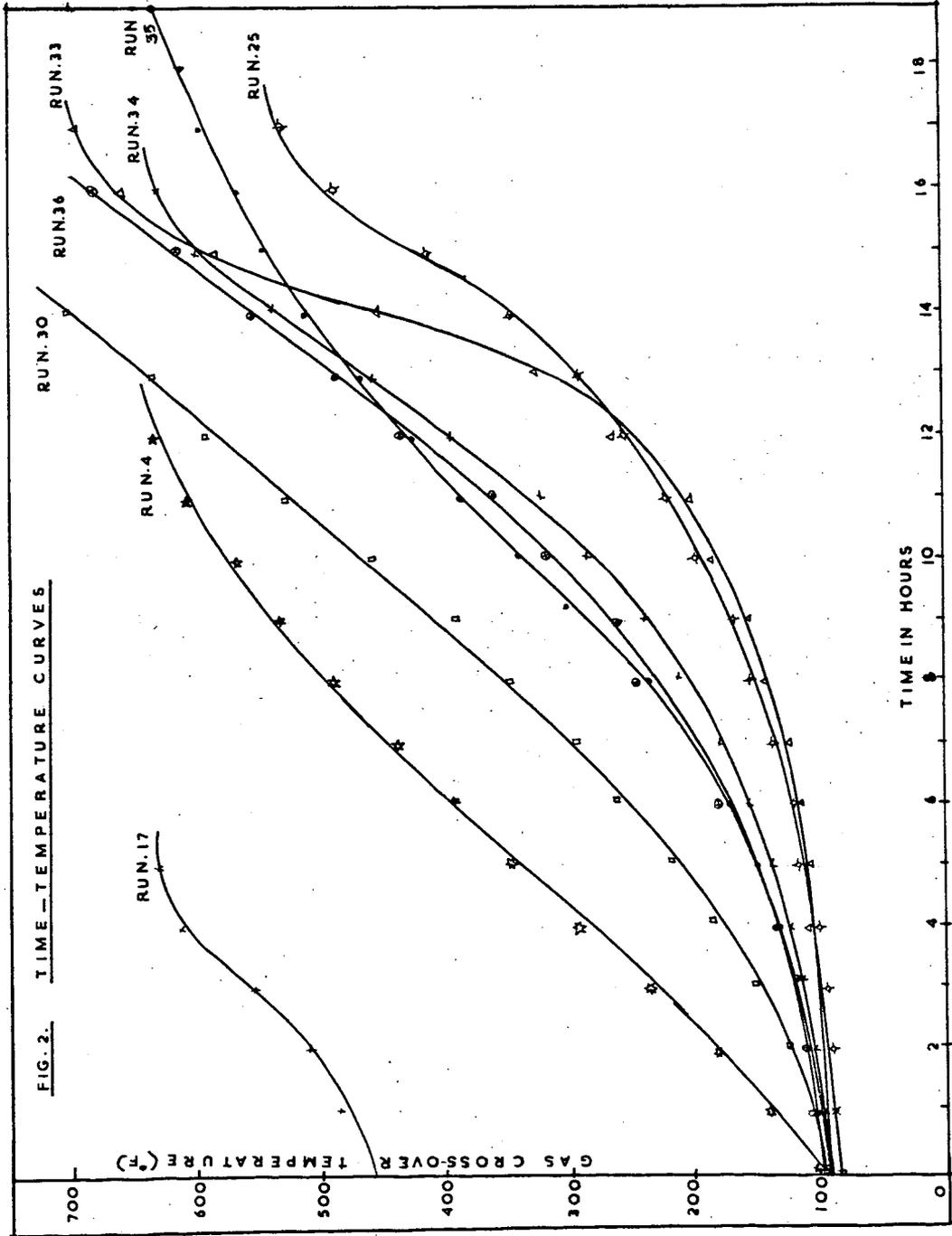
Run Nos.	30	17	20	4	25*	33
<u>Raw briquettes:</u>						
Moisture %	12.4	12.0	12.0	12.7	13.2	11.4
Ash %	5.9	7.5	8.2	8.9	9.4	14.4
Ash (Moisture free)	6.7	8.5	9.4	10.4	10.7	15.5
Shatter index + 3/4" %	87.5	90.3	93.5	86.7	86.2	84.8
Tumbler index + 3/4" %	82.6	76.1	72.4	67.6	57.4	62.5

*Low tumbler index in this run is due to the cumulative effect of high moisture and ash.

TABLE 7 - PROPERTIES OF RAW AND CARBONISED BRIQUETTES

	Raw briquettes (South Arcot lignite)	Carbonised briquettes (South Arcot lignite)	Charcoal (6)	Lignite char from Boehlen (5)
Moisture	% 11.5 - 12.5	1.0 - 3.5	4.54	3.1
Ash	% 5.9 - 8.2	11.2 - 15.8	4.55	24.4
Volatile matter	% 40.6 - 44.2	13.5 - 15.6	13.01	11.8
Fixed carbon	% 35.4 - 40.5	70.8 - 73.6	77.90	60.7
Calorific value	Kcal/kg. 5120 - 5650	6250 - 7020	7100	6167
Shatter test + 3/4" %	89.2 - 94.6	91.0 - 96.2	82.40	-
Tumbler test + 3/4" %	78.2 - 82.6	74.8 - 80.2	70.3	-





NOTE ON CHARGING TEMPERATURE AND COKE QUALITY
REPRESENTATIVE STUDY RESULTS OF AMERICAN, AUSTRALIAN AND
JAPANESE COALS

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1. Introduction

During the past few years, the Coal Research Group of the Department of Geology and Geophysics, University of Sydney, has been engaged upon studies designed to assist in the better development and more economic utilisation of the solid fuels. One of the most comprehensive fields of investigation in which studies are still proceeding, is that of carbonisation; the principal concern has been to investigate the factors affecting carbonisation and to improve coking characteristics in relation to metallurgical coke production. The seams studied to date include both established coking coals and some generally regarded as poorly coking and unsuitable for industrial use; they have been drawn from American, Australian and Japanese coalfields.

In 1955, the senior author, with a group of American co-workers from the Illinois State Geological Survey, carried out an intensive laboratory-scale investigation of the petrographic and coking characteristics of the Illinois No. 6 and No. 5 coals (1). Subsequently, the Sydney research group embarked upon an extended project of utilisation studies of some Australian seams which are of similar character to the Illinois coals; interim statements of certain early results were submitted to the Institute of Fuel (Australian Membership) Symposium at Newcastle, N.S.W. in 1959 (2,3). More recently the project has been greatly expanded to cover a wider range of both individual and blended coals, particular emphasis being placed upon the blending potential of selected Australian and Japanese seams.

In all this work, it is recognised that the laboratory scale study results cannot be integrated directly with those of full scale industrial practice, but experience has shown that they may be correlated. Consequently, the laboratory methods permit a relatively rapid, economic and critical assessment of the factors affecting coking characteristics, and establish trends which may serve as useful guides to improved industrial practice.

2. Coking Potential

Broadly, the many related and mutually modifying factors which appear to affect the coking potential of a coal may be grouped as (a) inherent petrological, physical and chemical characteristics of the raw material; (b) induced modifications of these characteristics brought about in the preparation of the oven charge; and (c) conditions under which the coal is carbonised. Within and between these groups all factors are subject to mutual variation; consequently critical assessment of coking potential demands comprehensive, exhaustive and controlled studies.

The present paper deals only with one limited aspect of the influence of carbonisation conditions upon coke characteristics, namely, the effects of charging temperature upon the mechanical properties of the resultant coke. Other conditions and standards adopted for these tests have been based upon much more comprehensive studies, discussion of which is beyond the scope of the present paper.

However, it should be stated that detailed studies have shown that coal particle size consist is often a very significant factor in determining the physical qualities of the coke produced. Under the conditions of study developed, the majority of normal, medium to high-volatile bituminous coals which have been investigated to date appear to yield most satisfactory cokes when the raw material is reduced

by controlled breakage to pass $\frac{1}{8}$ " mesh, with the minimum production of finer sizes. Standardised procedures of size reduction, however, do produce differences in final consists, a response to variations in the physical and petrographic characteristics of the seam material.

3. Coke Production and Testing Methods

The methods of laboratory-scale production testing and evaluation developed for the coking studies of the Illinois seams have been described elsewhere (1), and compare closely with those more recently applied to similar investigations of the Australian and Japanese coals. For the purposes of the present study, coal charges of approximately 700 grams, prepared according to a standard procedure, were coked under various conditions of charging temperature, final soaking temperature, and length of coking period. The coal charges, contained in closed, cylindrical refractory retorts, were heated in a silit-rod (Globar) type of electrical furnace with accurate thermostatic and program control. All coke runs were made in quadruplicate. The "strength" characteristics of the resultant cokes were examined by laboratory scale adaptations of the industrial shatter, tumbler and micro-mechanical tests.

The combination of "strength" tests used was considered to be most suitable in the circumstances, as shatter and macro-tumbler stability index results are conditioned largely by the macro-structure of the coke (joints, cracks, pore-dimensions and "wall"-thickness) while micro-mechanical strength and macro-tumbler hardness largely indicate the strength of the coke substance itself. That these laboratory tests were adequately discriminatory is evident from the study results summarised in figures 1-8.

In the following discussion, "optimum" as referred to coke quality implies maximum attainable values for the mechanical strength indices. Unfortunately it is seldom that all these strength indices attain their respective maxima under precisely similar conditions. Consequently the "optimum" of quality in any coke must represent a compromise, most suited to the consumer's particular requirements. Similarly, "optimum" conditions of coking are described by those particular circumstances in the thermal cycle which appear to contribute to the development of the highest mechanical strength indices.

4. Factors in the Thermal Cycle of Coking

The factors considered to be of importance in the thermal regime include

- (a) Oven temperature at charging) particularly in relations to the plastic
- (b) Rate of heating) range of the coal.
- (c) Final coking temperature
- (d) Period of coking, both over all and at final temperature of carbonisation.

Limitations of space preclude any adequate discussion of all these factors although they are certainly important and mutually disturbing. However as an essential introduction to the main theme (the effects of oven temperature at charging upon mechanical strength characteristics of the coke produced) the following brief and qualified resumé of the general effects of the other factors in the thermal cycle is included.

Final Coking Temperature

For the majority of "normal" bituminous coals examined so far, increased final coking temperature is accompanied by improvements in coke quality in respect of both macro- and micro-mechanical indices, until a particular "optimum" temperature of coking is attained. Carbonisation temperatures above the "optimum" for each particular coal, invariably have resulted in severe deterioration in the macro-mechanical indices of the coke. In the laboratory studies, for most of the coals prepared under "standard" conditions, the particular "optimum" temperature of final carbonisation lies between 1000°C and 1200°C, although in some cases further improvement in mechanical strength may be induced by even higher temperatures.

In the case of the Illinois coals, "optimum" final coking temperature proved

to be 1850°F (1010°C); for the Australian and Japanese coals, 1200°C has emerged as the most nearly satisfactory temperature of final coking.

Duration of Coking

The majority of the coals studied appear to yield "optimum" cokes when "soaked" at the final coking temperature for a relatively short period. In the laboratory studies, periods of more than two hours at maximum temperature of carbonisation were usually accompanied by definite deterioration in the coke strength, although this invariably improved again with further extension of time to approximately 10 hours. As such prolonged "soaking" was not considered practicable, the two hour final soaking period was adopted as standard.

Under "standard soaking" conditions, the coke produced usually exhibited a "steely", bright lustre and a minimum of breeze.

Rate of Heating

In coking, rate of heating must be related to the initial temperature and heat capacity of the oven on charging, the oven temperature increment per unit of time, and the thermal conductivity of both the coal charge and the progressively developing zone of coke, quite apart from the thermal characteristics of any reactions involved.

The influence of oven charging temperature upon coke-strength characteristics is discussed later as the principal theme of this paper. It is sufficient to observe here that the higher the oven temperature upon charging, the more rapid is the initial rate of heating, a factor which appears to be of particular importance in relation to the plastic temperature range, plasticity and volatile evolution characteristics of the coal.

In the laboratory studies, the initial rate of heating of the charge was increased by introducing the coal into the oven at successively higher temperatures. Thereafter, dependent upon the characteristics of the equipment available, standard rates of temperature increase were maintained at 3.6°C/min. (American coals) and 3.4°C/min. (Australian and Japanese coals) to 1000°C, followed by slightly reduced rates of heating to the maximum "soaking" temperatures. These figures closely approximate mean rates of temperature increase established in pilot oven studies, and were accepted as "standard" but subject to later investigation.

5. Charging Temperature and Coke Quality

Representative analyses of the American, Australian and Japanese coals which formed the subject of this study are quoted in Table 1.

TABLE 1 - Analyses of Coal

Seam	Prox. An. (% a.d. coal)				Ult. An. (% d.a.f. coal)					B.S. Index
	Moist.	Ash.	Vol.	F.C.	C.	H.	N.	S.	O.	
No. 6 Coal, Illinois	7.8	8.6	34.8	48.8	80.8	5.5	1.9	-	-	4
No. 5 Coal, Illinois	5.2	10.9	34.4	49.5	82.3	5.5	1.9	1.6	8.7	3½
Liddell Coal, N.S.W.	3.0	8.0	36.8	52.2	82.4	5.8	2.3	0.4	9.1	2½
Borehole Coal, N.S.W.	2.5	9.9	32.6	55.0	82.4	5.4	1.7	0.5	10.0	5
Bulli Coal, N.S.W.	1.1	7.2	22.2	69.5	89.0	5.0	1.9	0.3	3.8	8
Akahira, Japan	2.8	5.5	41.8	49.9	82.3	6.2	2.3	0.6	8.6	5
Futase, Japan	3.0	8.4	38.2	50.4	82.3	6.0	1.5	0.3	9.9	1½
Takashima, Japan	2.2	5.9	43.6	48.3	83.8	6.3	1.4	0.6	7.9	7

Illinois seams Nos. 5 and 6, the Liddell and Borehole of New South Wales, are quite comparable in chemical constitution and similar in overall petrographic character; the Bulli coal of New South Wales and the three Japanese samples were respectively of somewhat higher and lower rank.

Standard charges of each of these eight coals (controlled bulk and size consist) were introduced into the coking oven, of which the initial temperature was increased by successive increments to the particular final coking temperature adopted as optimum for each coal. In all cases, after charging, the oven temperature increase was maintained at the standard, controlled rate until the soaking temperature was attained.

With the exception of the Bulli and Takashima coals, all those examined revealed generally similar trends in the relationship between coke strength and oven charging temperature. In general, the majority of the strength indices improved progressively, reaching maxima for various particular charging temperatures ranging between 400°C and 1000°C; the decline in the strength index beyond the maximum was frequently precipitate. In the case of the Bulli coal, increased charging temperatures produced an initial deterioration in the strength indices of the coke, which thereafter improved to their particular maxima before declining rapidly. The Takashima coal in general exhibited deterioration in macro-strength indices and improvement in micro-strength characteristics with increased charging temperature.

Though frequently having pronounced effects upon the other strength characteristics, the temperature of oven charging assumed most critical significance in relation to shatter and stability indices - both measures of the coke macro-structure. Severe deterioration of one or both of these indices is not infrequently accompanied by slight improvements in the strength of the coke substance, as is made evident by increased micro-strength and resistance to abrasion.

(a) The American Coals

The most significant feature of these study series, results of which are shown graphically in Figures 1 and 2, was the critical effect of charging temperatures in excess of 450°C and 540°C respectively on the shatter and stability indices. Progressive increases in charging temperature from 25°C to these critical values were accompanied by modest though erratic general improvements in shatter and some reduction in stability for the No. 6 coal; a general, gentle, but varying deterioration in all macro-strength indices occurred in the case of the No. 5 coal.

When charging was carried out at temperatures progressively higher than those indicated, both shatter and stability indices of the cokes produced from each of these coals exhibited a general and rapid decline. In each case the deteriorations were accompanied by slight improvements in resistance to abrasion and micro-mechanical strength, the trends being more strongly marked in the higher ranges.

It appears significant that in each case the critical charging temperature (450°C for No. 6 coal and 540°C for No. 5 coal) is within or just adjacent to the plastic temperature range of the coals. It is considered that when charged at successively higher temperatures, these coals are subjected to an ever-increasing rate of temperature increase up to, through, and above the plastic range, and are thus afforded less and less time for volumetric and other adjustments before the "setting" temperature is reached. The consequent accumulation of stresses and the development of increased jointing and fracturing is indicated by the decrease in macro-mechanical strength.

The toughness or hardness of the coke substance (resistance to abrasion and micro-mechanical strength) does not appear to be related to the heating rate in any particular range, and increases slowly with the rise of charging temperature.

Apart from the effects upon strength, increases in charging temperature were accompanied by marked and progressive improvement in the colour and lustre of the resultant cokes, which were also of rather smaller size consist.

(b) The Australian Coals

As in the case of the American coals, charging temperature again assumed particular and critical significance in relation to coke shatter and tumbler stability indices, but serious and progressively increasing impairment of these factors did not occur until it exceeded 800°C. Unlike the Illinois seams, there was no

serious and continuing decline in macro-strength of the coke produced, with charging temperatures immediately above the coal plastic range. However, the Liddell coal (Fig. 3) did exhibit a slight depression of all three macro-mechanical strength indices associated with a charging temperature of 500°C which is slightly above the setting point. Progressive increases in charging temperatures to 800°C were accompanied by definite improvements in the macro- and micro-strength indices of the coke produced. Higher charging temperatures induced serious deterioration in macro-strength.

The Bulli coal (Fig. 5) returned a severely weakened coke when charging was carried out at temperatures of 200°C (very low macro-tumbler indices) and 400°C (depressed shatter and micro-mechanical strength); charging at increasing temperatures between 400°C and 800°C produced general and progressive improvements in the coke strength with well defined deterioration thereafter. Optimum micro-strength was obtained from coke produced by charging at air temperature. The plastic range of Bulli coal is approximately 390°C to 480°C.

The Borehole coal showed continuous and progressive improvement in all macro-strength indices of coke produced with increase of charging temperature up to 800°C (Fig. 4); thereafter shatter index declined abruptly although tumbler stability did not deteriorate until the charging temperature exceeded 1000°C. Charging temperatures above 800°C were accompanied by accelerated improvement in coke micro-strength.

In each of these coals, the higher charging temperatures were accompanied by considerable improvement in both colour and lustre of the coke produced.

(c) The Japanese Coals

For the three Japanese coals, variations in charging temperature produced three generally quite different trends in overall and particular variation of coke strength. The Akahira coal behaved in a manner broadly comparable with the Illinois coals Nos. 5 and 6; the Futase revealed trends rather similar to those of the Borehole coal of New South Wales; the Takashima coal could not be readily compared with any other coal as yet studied.

The Akahira was the only one of the eight coals discussed here which did not show an essentially parallel trend of shatter and stability indices throughout the greater part of the charging temperature range. The results of the macro-mechanical tests of the Akahira cokes suggest that a progressive increase in charging temperature to 1000°C brings about the formation of a joint or fracture system in the coke which is progressively more susceptible to rupture from impact but progressively less susceptible to abrasion. Micro-strength indices improved slightly and erratically with increased oven temperature at charging.

In the case of the Futase coal (Fig. 7) all macro-strength indices improved with increased charging temperature up to 600°C; thereafter the shatter index declined rapidly, followed by tumbler stability for temperatures exceeding 800°C. Micro-strength variation was erratic and not evidently significant.

The Takashima coal yielded a coke of superior macro-mechanical strength if charged to a cold oven. Progressive increases in the initial oven temperature to 800°C brought about a very substantial and progressive decline in these characteristics, after which there was a slight improvement. A somewhat erratic but definite improvement in micro-mechanical strength is evident in response to increased charging temperatures.

Variations in the appearance and size characteristics of Akahira and Futase cokes corresponded closely with those of the American and Australian groups. The Takashima, however, showed no significant variation throughout the range in either friability or the fragmentary character of the coke yield; only lustre improved as higher charging temperatures were employed.

Summary and Conclusions

From this present brief review of as yet incomplete studies, it is evident that the program of temperature increase during carbonisation has a profound and possibly particular effect upon the mechanical properties of the coke produced from any "coking" coal. Optimum qualities of macro- and micro-strength are

seldom developed together under any single coking regime. Consequently, both coke producer and consumer must accept a compromise on quality characteristics, according to their particular requirements and the inherent limitations to controlled coking. On the other hand, it is evident that much may be achieved in the control of coke quality through careful selection and preparation of the raw coal.

For certain high volatile bituminous coals (e.g. Illinois Nos. 5 and 6, and Futase), charging to the oven at temperatures even modestly in excess of their plastic range may well induce serious deterioration in the macro-strength characteristics of the coke. Other bituminous coals may, with minor exceptions yield substantially improved cokes when charged to an oven of which the temperature is 300 or 400°C higher than the plastic range (e.g. Liddell, Borehole and Bulli). Some coals may yield cokes which apparently deteriorate progressively in macro-strength characteristics with any increase of charging temperature above that of an air-cold oven.

No significantly progressive deterioration in coke micro-strength characteristics has been observed as accompanying increased oven temperature on charging. On the contrary, in the majority of cases, the strength of the coke substance appears to improve with increased initial oven temperatures, particularly in the higher ranges.

It is suggested that variations in coke character may be significantly influenced by rates of heating as related to the plastic range, plasticity characteristics, and gas evolution characteristics of the coal concerned; these in turn reflect the petrographic constitution, type distribution and rank of the seam. Further work is proceeding in this field of investigation.

References

- (1) Marshall, C.E., Harrison, J.A., Simon, J.A. and Parker, M.A. 1958. Petrographic and Coking Characteristics of Coal: Laboratory studies of Illinois coal seams Nos. 5 and 6. Illinois State Geological Survey Bulletin 84.
- (2) Tompkins, D.K., 1959. Preliminary Studies of the Coking Characteristics of Liddell Seam Coal. Institute of Fuel (Australian Membership) Symposium on Australian Fuels and their Utilization: Symposium Preprint.
- (3) Branagan, D.F., 1959. Some Aspects of a Coking Study of the Borehole Seam. Institute of Fuel (Australian Membership) Symposium on Australian Fuels and their Utilization: Symposium Preprint.

Figure 1.

ILLINOIS No. 6

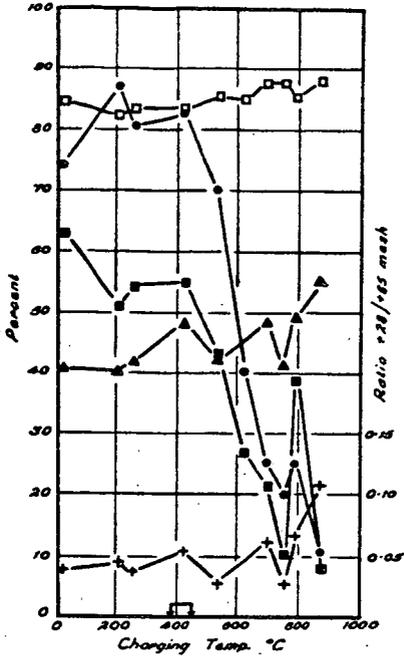
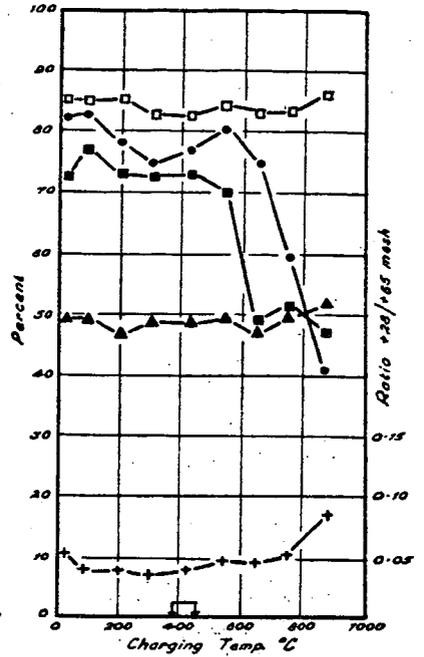


Figure 2.

ILLINOIS No. 5



LEGEND

- — ● +1" Shotter Index
- — ■ +1" Tumbler Stability Index
- — □ +1/4" Tumbler Resistance to Abrasion
- ▲ — ▲ Microstrength 65
- ✦ — ✦ Microstrength 28/65
- ⌞ — ⌞ Plastic Range

Figure 3.

LIDDELL

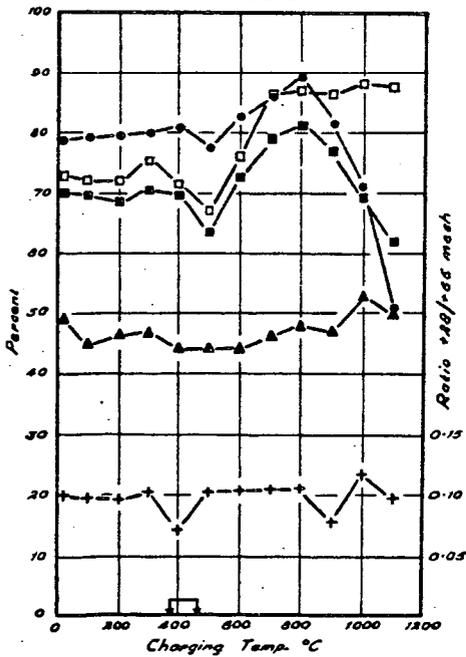


Figure 4.

BOREHOLE

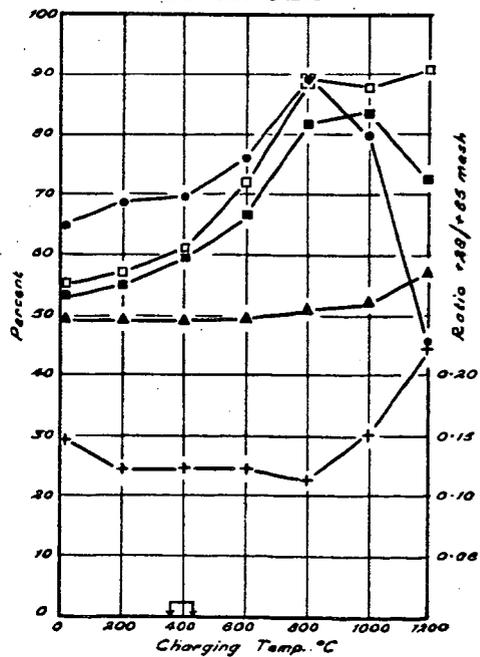


Figure 5.

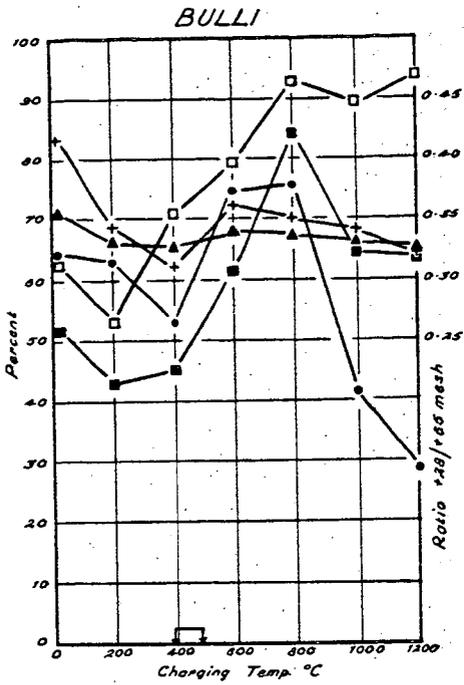
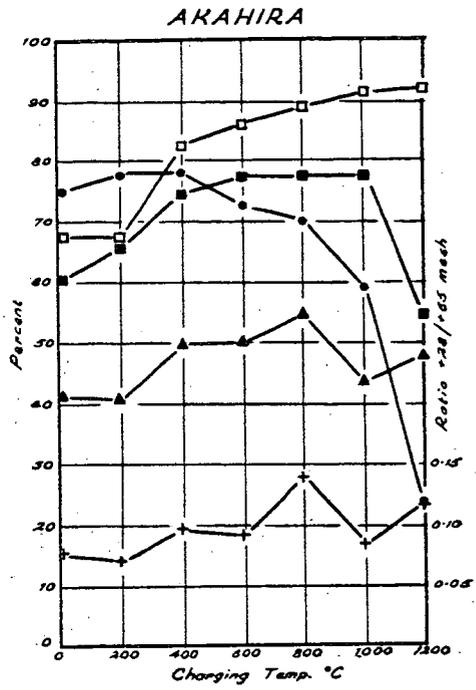


Figure 6.



For Legend, see previous page

Figure 7.

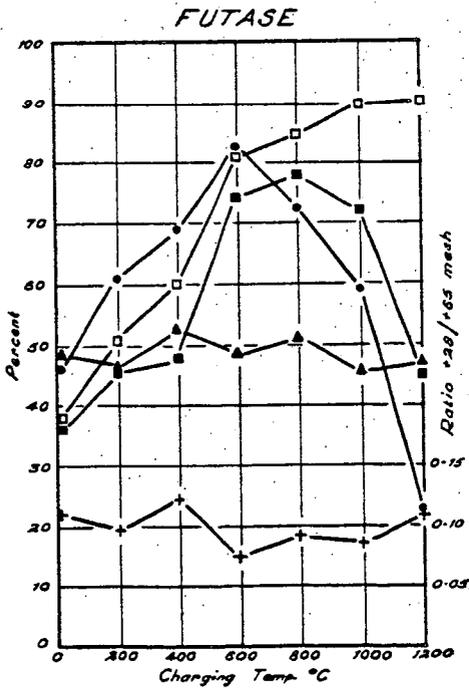
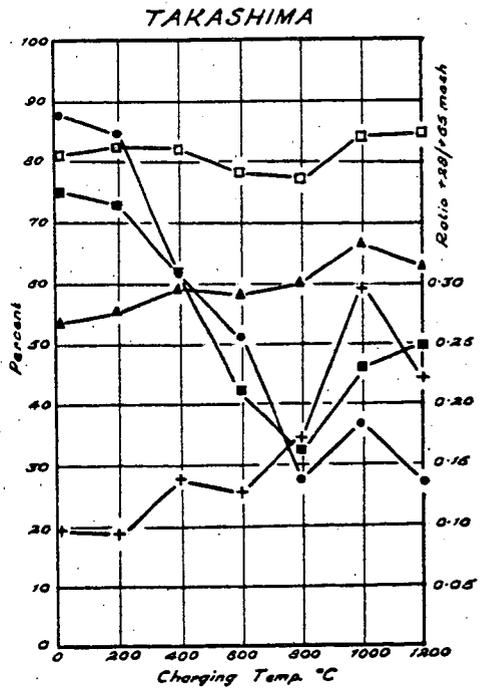


Figure 8.



RESEARCH INTO METHODS OF IMPROVING BLAST FURNACE COKE QUALITY
AT B.H.P. NEWCASTLE, AUSTRALIA
WITH SPECIAL REFERENCE TO THE EFFECT OF BENTONITE

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The high ash, medium to high volatile coking coals available in the vicinity of the Iron and Steel works at Newcastle⁽¹⁾ yield a very poor quality blast furnace coke by most world standards. This factor, combined with rapid expansion of the Company's production facilities, has emphasised the need for extensive investigation of methods of improving the quality of the coke made from these coals.

Fine grinding of the coal charged to the ovens at Newcastle has been of recognised practical benefit, and in fact, common plant practice for some thirty five or more years,^(2,3,4) and the grind specification over this period has generally varied between the equivalent of 50% to 70% minus 18 mesh B.S.S. which is very fine by most standards. However, with the expansion of the company's research activities and the installation of pilot scale research equipment⁽⁵⁾, it was considered desirable to investigate the effect of coal grind and the use of additives in more detail.

An early stage of this research comprised experimental and full scale oven tests on the effect of coal grind on coke strength. In addition, many series of tests comparing the effect of various additives previously investigated, such as char made from non-caking coals, were carried out over a range of coal charge sizings. In the course of the above investigations, observations made in relation to the effect on coke strength of Wyoming bentonite and the bentonitic clayshales associated with the unwashed coals resulted in the concentration of a good deal of research effort on mineral matter associated with both northern and southern New South Wales coal seams.

Considerable detailed research on the effect of additives such as char, coke-breeze, iron ore, and limestone has been carried out which fully confirms the advantages of char addition in comparison with other recognised methods of coke strength improvement. However, it is not intended to describe this work in any detail here, save to compare the coke strength data with that obtained in the investigation of finer grinding and bentonite.

EXPERIMENTAL OVEN TESTS ON UNWASHED COALS

In November 1956, as part of a long range program of improving the quality of blast furnace coke used at the Newcastle steelworks, investigations of the coking characteristics of the northern coal seams and variations between the same seams at our several collieries were commenced. These experimental oven tests were carried out prior to the installation of the Pilot Scale Coal and Coke Research Laboratory⁽⁵⁾, and consequently, close control of such charge variables as ash content and coal grind was not always possible. A large number of oven tests were of necessity carried out in an attempt to clarify the effect of the many variables encountered.

The results of a series of earlier experimental oven tests are shown in figure 1, in which the trend in relationship between coke strength indices and coal grind are shown.

It can be seen that increasing the fineness of grind of the coal charge has resulted in an increase in the strength of the coke as measured by the A.S.T.M. stability, hardness, and shatter indices. These results indicated the possibility of increasing the stability of the coke from less than 20 to over 30 by increasing the fineness of grind. Coke of 30 stability has previously only been obtained on a production basis at Newcastle by adding approximately 25% lower volatile southern N.S.W. strongly coking coal to the northern blends. The incorporation of this southern coal, although resulting in improved blast furnace operation, has generally been considered an expensive answer to the problem of improving Newcastle blast furnace coke quality. The results obtained by finer grinding suggested therefore, an alternative method to the use of southern coal. However, it was also realised that there existed a number of practical problems in relation to achieving and handling on a production scale grinds finer than 70% minus 18 mesh.

FULL SCALE FINE GRIND TESTS ON UNWASHED COALS

In February to April 1958, ten series of fine grind trials were carried out in the full scale ovens at Newcastle. The bulk of these tests were carried out using Lambton Colliery, Victoria Tunnel Seam, Burwood Colliery, Borehole Seam and Burwood Dudley Seam coals in the unwashed condition. Facilities were not available to carry out tests on individual seams after washing, but it was possible to test the effect of coal grind on the then current northern blend ex the washery and the final test in this series was conducted on such a sample washed to 14% ash. The overall result of the effect of coal grind on coke strength for this series of tests, on coals ranging from 13% to 21% ash content, is shown in figure 2. Calculation showed that these full scale oven tests gave a close correlation with the results obtained in the experimental oven on northern coals at a similar ash level.

Promising though these results were in regard to the effectiveness of finer grinding, the discovery of the beneficial effect of small percentages of bentonite and bentonitic shales in northern coals^(5,5), provided reason for caution in assessing the effectiveness of fine grinding. In addition, during the tests, considerable difficulty was experienced in handling the very finely ground coal charges, which is a further important consideration in regard to finer grinding.

A closer examination of the results of the full scale fine grind tests on Victoria Tunnel, Dudley and Borehole seam coals showed that there were at least three variables which could be contributing to the increase in coke strength, the other two being ash content and coal seam variation. Fine grinding had been most effective with the high ash Dudley seam coal, and least effective with the lower ash Borehole seam coal.

In addition, later investigations showed that of the coal seams tested, the Dudley and Victoria Tunnel seams containing bentonitic clay-shales gave the best results on finer grinding, whereas the Borehole seam containing relatively little bentonite gave the least improvement.

Because of the number of variables that could be contributing to the effectiveness of fine grinding, it was clear that the problem of significantly and economically improving northern blast furnace coke quality by finer grinding was more complex than originally thought.

EXPERIMENTAL AND FULL SCALE OVEN TESTS ON WASHED COALS.

(a) Experimental Oven Fine Grind Tests

In 1959 full scale wet washing of the fine coals on Deister tables replaced dry tables and the ash in the washed coal to the commercial ovens was reduced from 15% to 11%. Blast furnace operation benefited considerably from the reduction of 4% in coal ash, however, there was a fall-off in coke strength. In spite of the fact that the 4% reduction in ash gave several points increase in coke A.S.T.M. hardness index, the A.S.T.M. shatter and stability were reduced somewhat and there was a marked increase in the amount of spongy coke produced.

The opportunity was taken to carry out experimental oven fine grind tests on these current washed coals of 11% ash. Two series of tests were carried out, each on a ten ton sample from the full scale washery. As anticipated, finer grinding was somewhat less effective with these washed coals. However, it was also found that in the experimental oven, these washed coals gave considerably better quality coke at coarse coal grinds than did the full scale ovens. Although the reasons for this improvement were obscure, it was clear that reasonably strong coke could be produced in the experimental oven at (by our standards), relatively coarse coal grinds of the order of 40% to 50% minus 18 mesh B.S.S.

(b) Full Scale Oven Tests Comparing Fine Grind, Char and Bentonite

Since mid 1956, some 850 experimental oven charges of approximately 850 lbs each had been coked in investigation of various measures taken to improve coke quality, and it had been concluded that the most promising avenues of further investigation were finer grinding, 5% char addition and 1% Bentonite addition. These three methods although presenting their own individual problems as regards full scale application, were considerably more effective in the experimental oven than say, iron ore, coke breeze or limestone additions.

Therefore, a number of full scale oven charges were prepared at the Works Research Laboratory (as described in more detail in the next section), in order to confirm the results obtained in the experimental oven. The results of these tests are given in table I.

TABLE I.
Full Scale Oven Tests Comparing Fine Grind,
Char and Bentonite Addition.

<u>Description of Charge</u>	<u>Coal Grind</u> <u>±-18B.S.S.</u>	<u>A.S.T.M. Coke Physical Test Data</u>		
		<u>1½ Shatter</u>	<u>1" Stability</u>	<u>1" Hardness</u>
<u>3 Control Tests</u> (Range)	40/55	68/74	16/24	68/71
<u>Mixed Charge</u> (No additions)	48	71	19	68
<u>1% Bentonite</u>	48	76	34	72
<u>5% Char</u>	44	82	38	68
<u>Fine Grind</u>	65	77	29	73

From Table I it can be seen that the experimental oven findings were confirmed. Although 1% bentonite addition was not the most effective, it was decided to carry out further tests with smaller percentages of bentonite. If sufficiently effective in improving coke strength, less than 1% bentonite addition would offer a simple method of carrying out full scale blast furnace trials with stronger coke to determine the value of the latter in relation to operation of our Newcastle blast furnaces.

EXPERIMENTAL AND FULL SCALE OVEN TESTS ON THE EFFECT OF BENTONITE ON COKE QUALITY

The discovery of the beneficial effect of a small percentage of bentonite on Newcastle coke strength was made in May 1953 in the newly installed Pilot Scale Coal and Coke Research Laboratory (figure 3), and became the first major research program using this equipment. As has been reported elsewhere (5, 6, 7), a considerable number of experimental oven tests (figure 4) were carried out in investigation of the effect of both Wyoming bentonite and the bentonitic shales associated with the Newcastle coal seams.

From these experimental oven tests, it was shown that, although variable in properties, the local bentonitic clayshales associated with the coal seams approached Wyoming bentonite in their capacity to affect an improvement in the strength of Newcastle coke. It was also shown that 1% bentonite added to the lower volatile strongly coking southern N.S.W. coals had a marked detrimental effect on coke quality.

When the opportunity arose to carry out full scale oven tests to confirm these effects, it was decided to concentrate initially on the use of Wyoming bentonite in order to keep the test variables to a minimum.

(a) Full Scale Oven Tests Prepared at the Works Research Laboratory in a Spiral Mixer

The first full scale oven test with 1% of Wyoming bentonite added to the coal was carried out in March 1960 and resulted in such a marked improvement in coke quality that attention was concentrated on the addition of only 1/4% to 1/2% of bentonite. These tests were carried out by withdrawing full oven charges from the battery bunkers using the spare charging machine. After charging the necessary control tests, a full oven charge was transferred to a lorry at ground level and taken to the pilot plant laboratory where the dry bentonite was carefully blended using a spiral ribbon mixer. The mixed charge was bagged and returned to the battery where the four hundred bags (approx.) were hoisted to the top of the battery and emptied into the canisters of the charger. The results of the physical tests carried out in triplicate on the cokes produced in this manner are given in Table 2.

TABLE 2.

FULL SCALE OVEN TESTS WITH BENTONITE

<u>Description of Charge:</u>	<u>Coal Grind</u>	<u>A. S. T. M. Coke Physical Test Data</u>		
	<u>4 -18B.S.S.</u>	<u>1 1/2" Shatter:</u>	<u>1" Stability:</u>	<u>1" Hardness:</u>
<u>6 Control Tests</u>	<u>40/55</u>	<u>69/74</u>	<u>16/20</u>	<u>68/70</u>
<u>(Range)</u>				
<u>1/4 Bentonite</u>	<u>48</u>	<u>76</u>	<u>34</u>	<u>72</u>
<u>1/2 Bentonite</u>	<u>54</u>	<u>77</u>	<u>37</u>	<u>71</u>
<u>2/3 Bentonite</u>	<u>41</u>	<u>77</u>	<u>31</u>	<u>70</u>

From table 2 it can be seen that a marked improvement in coke strength has resulted from addition of either $\frac{1}{2}\%$, $\frac{1}{4}\%$ or 1% bentonite. The run-of-oven coke produced at Newcastle from the current washed coal of approx. 11% ash and 39% volatile matter (d.a.f.) besides giving poor coke strength, of the order of that shown for the control tests in the tables, contains an excessive amount of sponge coke. The addition of even a $\frac{1}{4}\%$ bentonite resulted in virtually complete elimination of this sponge and a less finery coke product. Figures 5 and 6 illustrate the difference in appearance of the cokes produced with and without bentonite addition. These photos are of a single bag of each coke selected at random. Even in a casual examination of the cokes, there could be little hesitation in selecting the one containing bentonite.

(b) Full Scale Oven Tests Prepared at Coke Oven Hammer Mills with no Separate Mixing.

The next series of full scale oven tests were aimed at determining the simplest practical method of adding the bentonite to the coal in anticipation of a full scale test in the blast furnaces over an extended period. It was decided to test the effectiveness of the hammer mills alone in mixing the bentonite with the coal. A small hopper and vibrating feeder were set up to feed bentonite on to the stream of coal feeding into the hammer mills. For the tests, sufficient coal was run through the mill to fill a sample bin (3 oven charges). Two or three control charges without bentonite were prepared and charged, followed by three charges with $\frac{1}{2}\%$ to $\frac{1}{4}\%$ dry bentonite powder added to the coal stream.

The results of the first test series carried out by adding the fine dry bentonite at the hammer mill are given in table 3.

TABLE 3:

FULL SCALE OVEN TESTS ADDING $\frac{1}{4}\%$ BENTONITE AT THE HAMMER MILLS

<u>Description of Charge:</u>	<u>A. S. T. M. Coke Physical Test Data:</u>		
	<u>$\frac{1}{2}$" Shatter:</u>	<u>1" Stability:</u>	<u>$\frac{1}{4}$" Hardness:</u>
Control	65	13	72
"	64	15	73
$\frac{1}{4}\%$ Dry Bent.	73	24	71
"	67	24	72
"	68	24	72

From the results of the above test, it was apparent that the coal used was of poorer quality than normal as reflected in the physical tests on the control charges. The test was repeated using the same technique of adding the bentonite at both $\frac{1}{2}\%$ and $\frac{1}{4}\%$ levels of addition. The results of these test series are given in table 4.

TABLE 4

FULL SCALE OVEN TESTS ADDING 1% AND
1/2% BENTONITE AT THE HAMMER MILLS

<u>Description Of Charge:</u>	<u>A. S. T. M. Coke Physical Test Data:</u>		
	<u>1 1/2" Shatter:</u>	<u>1" Stability:</u>	<u>1/2" Hardness:</u>
Control Tests	65	17	72
" "	63	16	70
" "	67	16	73
1/2 Dry Bent.	75	27	71
" "	70	30	75
" "	73	24	72
Control Tests	65	17	72
" "	65	18	74
" "	63	17	71
1/2 Dry Bent.	70	29	71
" "	75	30	70
" "	67	30	71

These full scale oven tests (table 4) confirmed that 1% to 1/2% bentonite could be added to the stream of coal entering the hammer mills with results comparable to those obtained by careful mixing.

DISCUSSION

Based on the results obtained in experimental and full scale oven tests, it is planned to carry out trials in the coke ovens and blast furnaces at Newcastle over an extended period. The Newcastle plant has a nominal capacity of 300 tons/hour of washed coal. We will aim at 1/2% dry bentonite addition to the coal stream entering the hammer mills. That is, we will be adding 1 ton/hour of ash forming material to the coal, and it must be admitted that the benefits obtained need to be sufficient to justify such an unusual precedent.

However, there is no doubt that bentonite markedly improves Newcastle blast furnace coke quality. We therefore, have the opportunity of answering the age old question concerning the benefits of coke quality on blast furnace operation, by a simple practical process. Undoubtedly, valuable information will be obtained.

Our aim as regards the coke strength in the blast furnace trial will be to produce a sponge free coke of the order of 75 shatter (1 1/2"), 30 stability and 70 hardness. Whether we can achieve this specification will depend to a large extent on the degree of control we can exercise over the uniformity of addition of the bentonite to the coal stream entering the hammer mills, and on the coal grind obtained in the mills. In most of the full scale oven tests carried out to date, the coal grind has been below 50% minus 18 mesh B.S.S. which is generally considered coarse for our types of coal. We know from experience that the beneficial effects of finer coal grind and bentonite addition can be combined to give a stronger coke. During the trial therefore, we will aim to maintain the coal grind above 50% minus 18 mesh (the minimum acceptable figure under normal conditions), which should ensure our achieving the desired coke strength indices.

There are many questions in our minds regarding the possible significance of the effect of bentonite on coke strength which can only be answered by further research and the proposed full scale trial.

On the fundamental side, there is the question of the mechanism by which bentonite affects an improvement in the quality of the high volatile northern N.S.W. coals. At the moment, our thinking favours the theory that the ion-exchange capacity of the bentonite favourably effects the stability of the bonding constituents in these coals. On the other hand, we have the contrast in effect on the lower volatile southern N.S.W. coals which in a limited investigation, have shown a marked deterioration in coke strength with bentonite addition.

The soft clayshales associated with the unwashed northern N.S.W. coals have been shown to possess bentonitic properties to varying degrees, some to a marked extent. The addition of 1% of bentonitic clayshale removed from the unwashed coal before washing and re-added to the washed coal has also resulted in a marked increase in coke strength. However, it is apparent that the beneficial properties of these clayshales are lost during coal washing, because after washing to 11% ash, these coals in practice, should contain sufficient of this clayshale to give an increase in coke strength. That an increase in coke strength has not resulted in practice is attributed to loss of ion-exchange capacity of the clays during washing. This aspect offers a further interesting field of research.

There remains something to be learnt regarding the best method of adding bentonite to the coal. A limited amount of work has been carried out on the addition of the bentonite in slurry form in both water and oil. The bentonite can be dispersed readily in the oil used for bulk density control, but it was considered that in this form, the ion-exchange capacity might be lowered to such an extent by the oil that the effect on the coke strength would be less marked. Results obtained to date with oil dispersion have been variable and further research is required on this point.

Water slurries of bentonite have given much the same order of improvement in coke strength as the dry bentonite additions. However, although by no means proven conclusively as yet, it is our impression that the moisture content of the coal may play an important role in the effectiveness of the dry bentonite addition. That is, the coal charges with higher moisture seem to give stronger coke with the dry bentonite addition. However, this issue is somewhat clouded by other variables at the moment.

On the production side, we hope to obtain an answer to the question of the relative merits of cokes of 30 stability and 15 stability in the blast furnaces at Newcastle. If the trial is successful from the point of view of control and coke strength and little effect is shown on blast furnace operation and production, then we will need to revise our assessments of what constitutes blast furnace coke quality. On the other hand, should blast furnace operation benefit significantly from the use of the bentonite coke, we will be a further step forward in the common goal of improving blast furnace productivity by the use of tailor made raw materials.

ACKNOWLEDGEMENTS

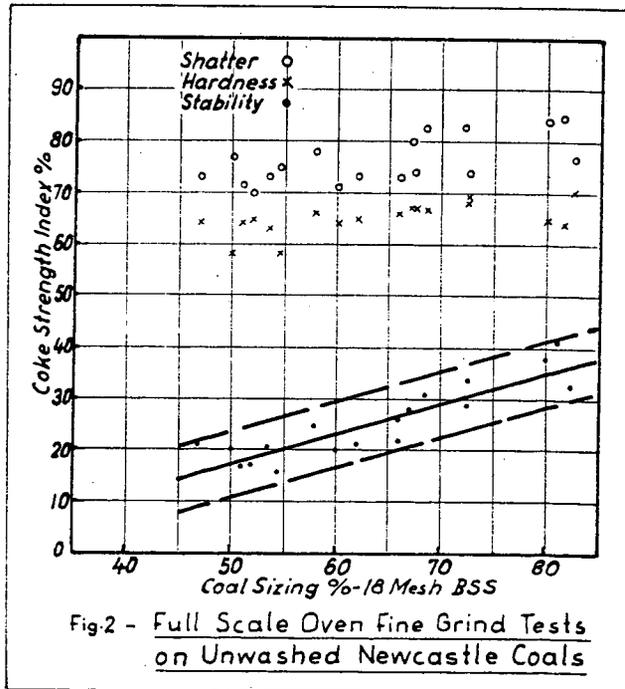
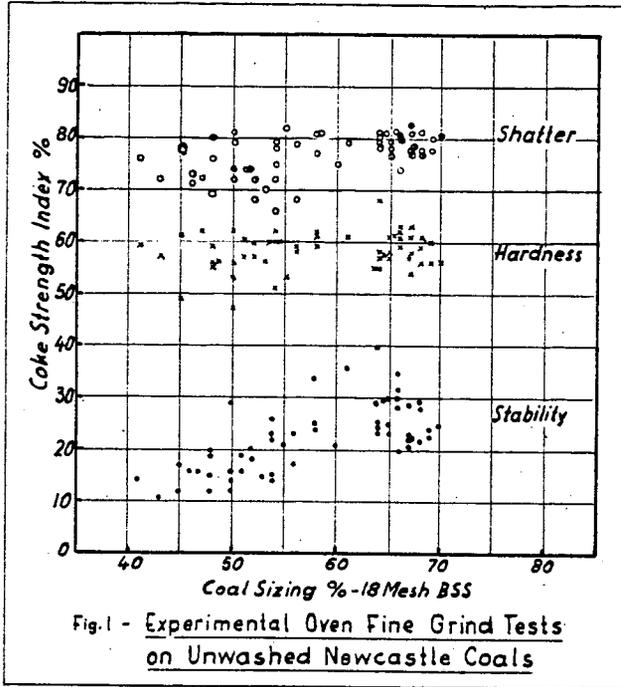
The author is indebted to the Directors and Management of the Broken Hill Pty. Co. Ltd., for permission to publish this paper.

The passage of this laboratory conception through the pilot plant stage to a full scale production trial required the cooperation and extra

effort of many colleagues together with constructive criticism at each step in the process. Although it is not practical to single out all of those who participated, the author is especially indebted to the Chief Works Research Officer, Mr. D.O. Morris and Works Research Officer, Mr. D.H. Felton who have been closely associated with this and every phase of our coal and coke research program.

REFERENCES

1. Standards Association of Australia - "The Coal Resources of Australia" Power Survey Report No.3 - 1955, p.29.
2. Butler K. - "Coal, Coke and By-Products - Their Treatment at Newcastle". B.H.P.Review, Vol.2, No.11, 23th Jan. 1925, p.5.
3. McLennan, I.M.- "Washed Coal as Applied to Metallurgical Practice" Bureau of Steel Manufacturers, Annual Meeting, 3rd Sept. 1940, p.40.
4. Norgard, J.D. & Brook, W.H. - "Coal Utilization in Modern Steelworks Practice". Preprint Aust.Inst.Mining & Metallurgy, May 1947, p.10.
5. Gregory, J.A. - "Pilot Plant Research into Methods of Improving Blast Furnace Coke Quality". B.H.P.Technical Bulletin, Vol.3, No.1, March 1959, p.23.
6. Gregory, J.A. - "The Effect of Mineral Matter in Coal on the Physical Strength of Blast Furnace Coke-Pilot Oven Test Results with 1% of Bentonite or Bentonitic Shale Added to the Coal Charge". The Institute of Fuel (Australian Branch) 1959, Symposium on "Australian Fuels and Their Utilization", Paper B11.
7. Felton, D.H. & Gregory, J.A. - "Pilot Plant Research into Methods of Improving Blast Furnace Coke Quality - A Summary of Experimental Oven Data". B.H.P.Technical Bulletin, Vol.4, No.2 1960.



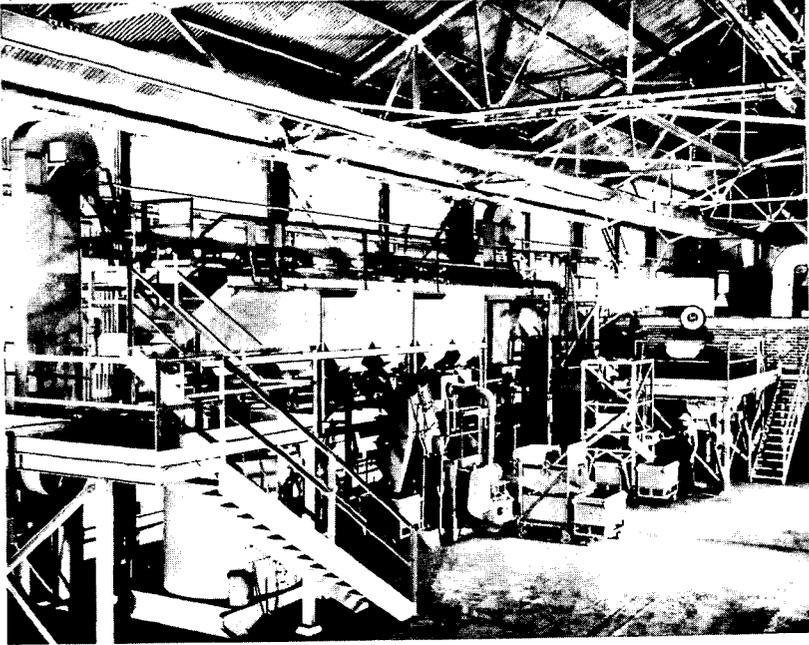


Fig. 3: - Section of the Pilot Scale Coal & Coke Research Laboratory.

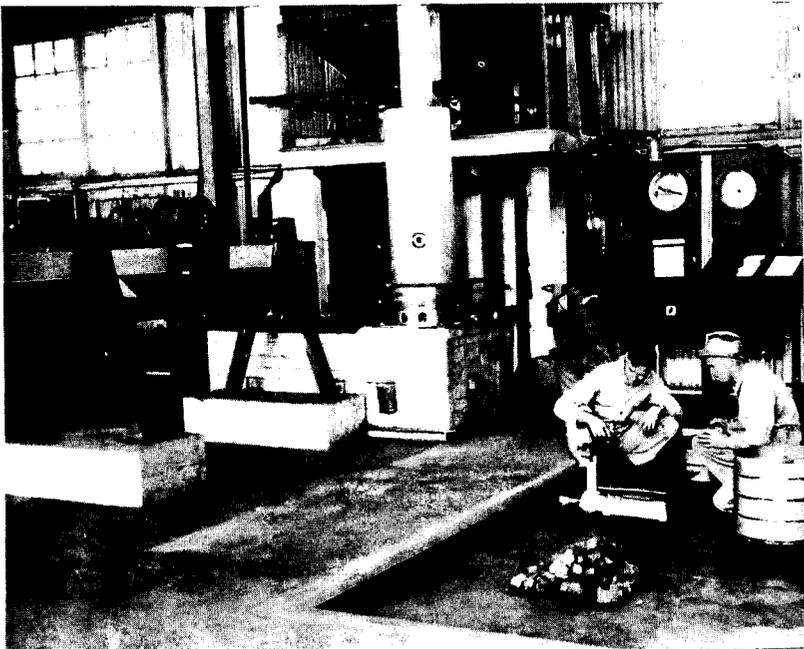


Fig. 4: - The Experimental Coke Oven.

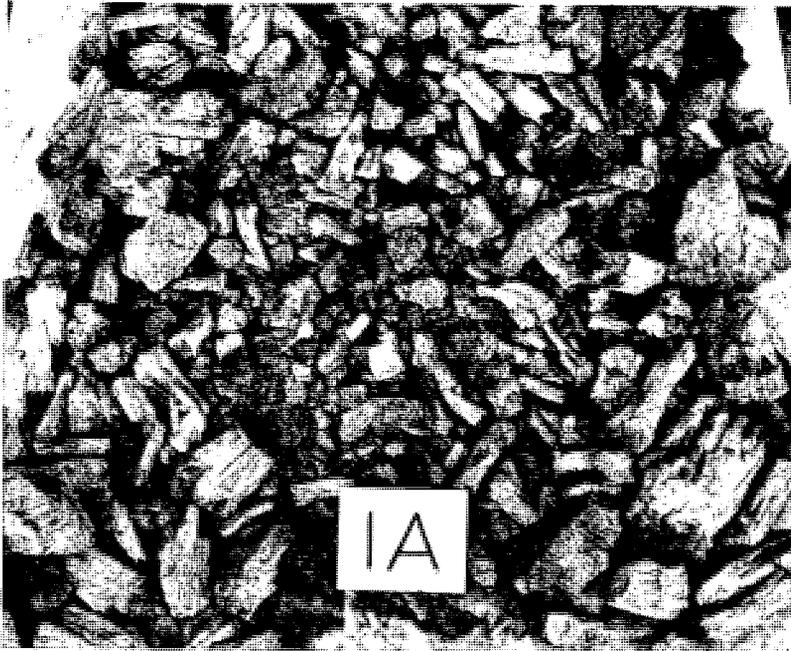


Fig. 5: - Control Test, Full Scale Oven, Typical Spongy Run of Oven Coke.



Fig. 6: - Full Scale Oven Test $\frac{1}{2}$ % Bentonite Addition.

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 ₂ O	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: 75%
(usually a mixture of various weakly coking coals)
- Coking coal 25%
(normally a mixture of coking coals).

A. THE PETROGRAPHIC COMPOSITION OF PREPARED COKING COALS.

The .../

* 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 vitrinerite 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 microscopic nodules of clay give rise to the typical high ash content of the South African coals.

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 $6\frac{1}{2}$ in the case of the lowest specific gravity fraction, to $4\frac{1}{2}$ 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-Arnu²⁾ and Hoffmann³⁾, as well as the Gieseler⁴⁾ 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 coke²⁾, 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 particles³⁾. 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 authors⁴⁾.

Some .../

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 Russel⁷⁾. 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/ft³.

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/ft³ for series 1 and 2, respectively. Those of the preheated coals were 54.9 and 49.5 lb/ft³, 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 work³⁾ 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.	Test 2.	Test 3.
	70% of A+30% of D normal crushing	As for Test 1 but A crushed to -1½mm	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 Blésbok coal. Fuel Research Institute of S.A. Technical Memorandum No.5/1958.
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SELECTIVE PREPARATION OF COALS FOR COKING*

by

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ABSTRACT

The selective preparation of coal for coking has been intensively studied during the past decade or so in various countries**; however, investigations on selective preparation were initiated more than 30 years ago***. Work on the Sovaco-Burstein process**** brought renewed interest throughout the world. Selective preparation is based on differential friability of the coal macerals with separation of enriched fractions of each by screening and recombining these in proportions that are optimum for the coals and coking process. Consequently, different coals present special problems in selective preparation. In this paper the results of work on Indian coals are presented. It would appear that selective preparation should prove beneficial to many of the coals of India.

* Manuscript not received in time for preprinting.

** C. W. Stahl, American Iron & Steel Institute Yearbook (1947) pp. 355-378.
M. S. Gryazhov, Lazguskii, I. L. and Feldbrin, M. G., Koks i. Khimiya (1956) No. 8, pp 3-10.
A. H. Brisse and H. E. Harris, Blast Furnace & Steel Plant, Vol. 45 (1957) March pp. 301-307.

*** K. Lehman: "Application of Petrography to Coal Preparation", Proc. of the Third International Conference on Bituminous Coal (1931) Vol. II. Carnegie Inst. of Technology, Pittsburgh, Pa., pp. 679-700.

**** R. Cheradame, Coke and Gas, April, 1954, pp. 147-149.