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

C. E. Marshall, D. K. Tompkins, D. F. Branagan, J. L. Sanderson

Department of Geology and Geophysics
University of Sydney,
N.S.W., Australia

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 particlar 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 overall 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

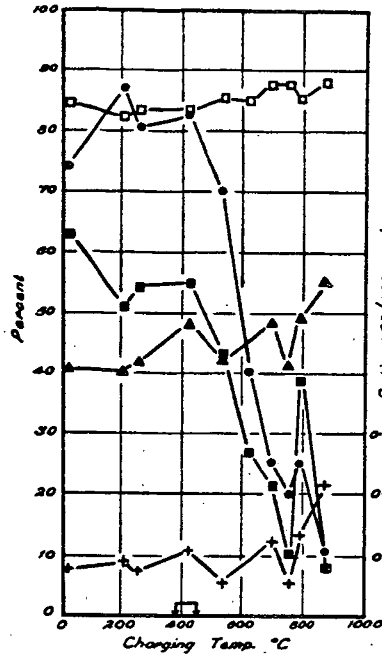
(1) Marshall, C.E., Harrison, J.A., Simon, J.A. and Parker, M.A. 1958. Petrographic and Coking Characteristics of Coal: Laboratory studies of Illinois coal seams Nos. 5 and 6. Illinois State Geological Survey Bulletin 84.

(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



LEGEND

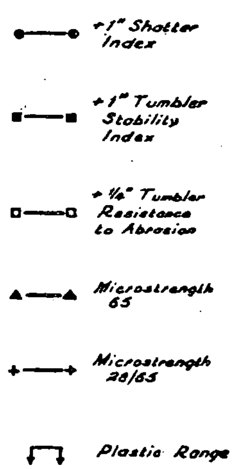


Figure 2.

ILLINOIS No. 5

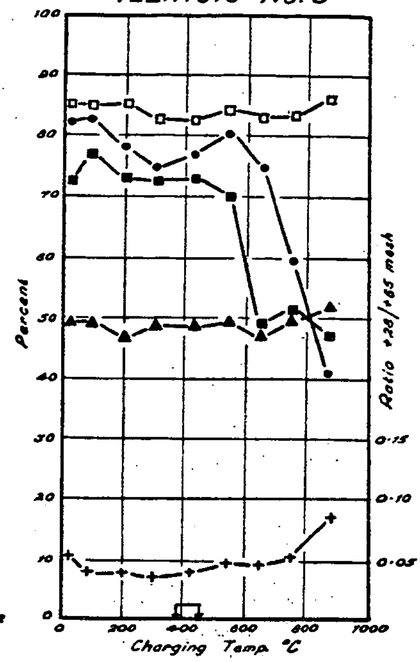


Figure 3.

LIDDELL

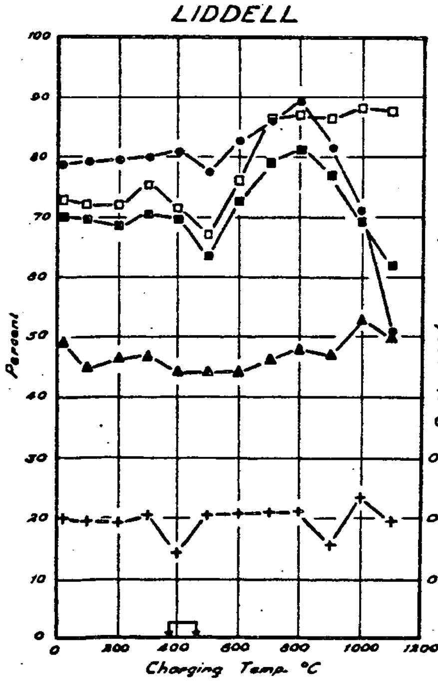


Figure 4.

BOREHOLE

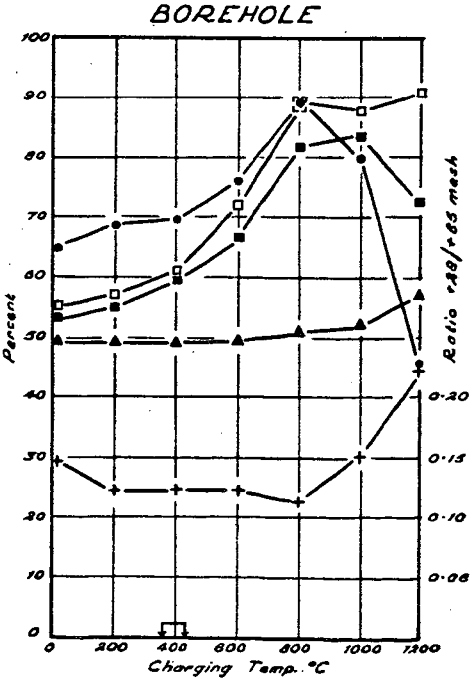


Figure 5.

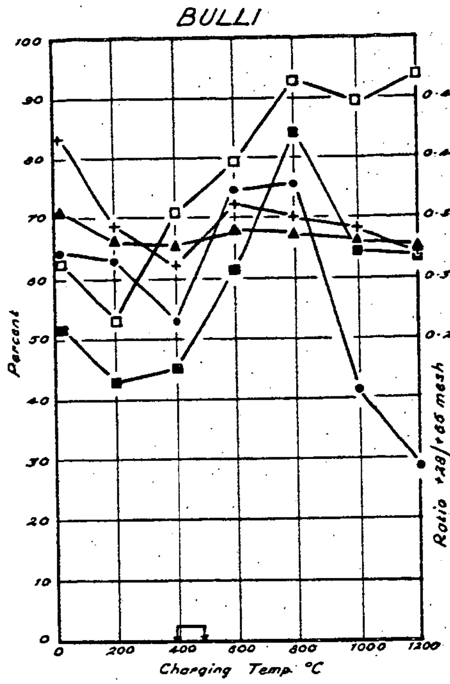
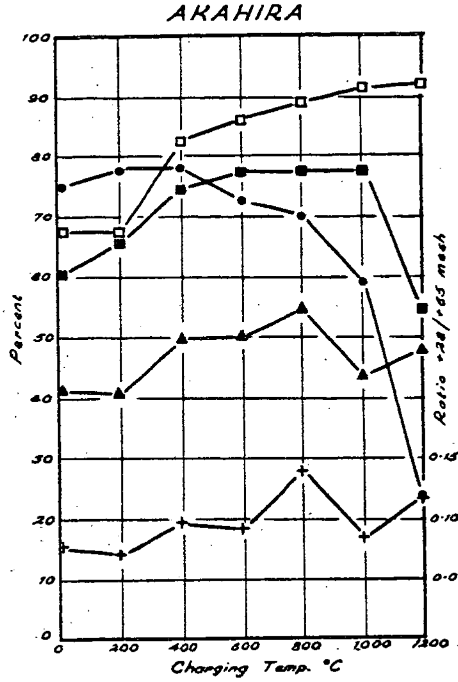


Figure 6.



For Legend, see previous page

Figure 7.

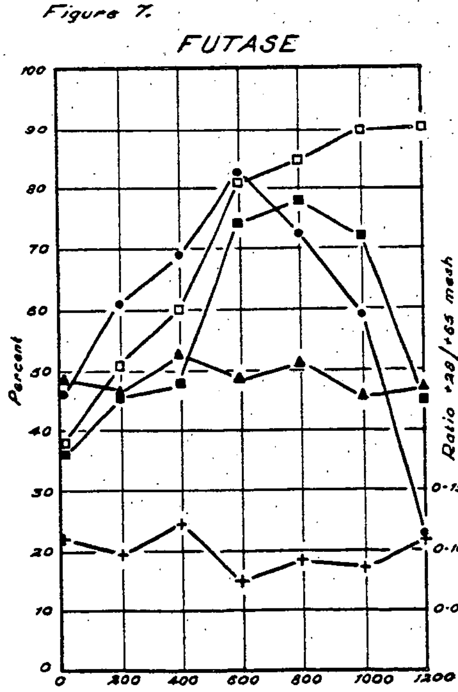


Figure 8.