

EVALUATION AND SELECTION OF COKING COALS

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This paper is based on a study and report by the Australian Coal Industry Research Laboratories Limited (ACIRL) by T.G. Calcott and O.C. Roberts. The support for the study was provided under the Australian Government National Energy Research Development and Demonstration Programme administered by the Department of Resources and Energy.

#### INTRODUCTION

Coals and iron ores are the primary raw materials of iron making after their conversion into coke and associated byproducts and sinter or pellets respectively. In the study, iron making operations are assumed to conform to the schematic presentation of Figure 1. This model permits the assumption that wharf coke is fully used in the iron making operations whereas if pellets were to be used instead of sinter, some separate account of breeze and nuts being less efficiently used would be needed.

Blast furnace cokes must possess physical and chemical attributes within certain limits to enable efficient and productive iron making. These limits vary from plant to plant. Coke size and strength limits are dictated by the requirement to provide adequate permeability to the blast while supporting the furnace burden. The practice of blending coals of different rank and caking properties is employed to achieve the desired physical properties in the coke. The chemical role of the coke

is to provide adequate amounts of heat, reducing gases and slag making components. Slag chemistry is regulated to control the composition of the liquid hot metal. Thus, the principal chemical attribute of coke is its carbon content, and it follows that the coke yields of the component coals of a coking blend have a large influence on iron making economics.

The properties of coals that determine coke chemistry also participate in determining the physical properties of blast furnace coke; but relations between the respective contributions are not simple. Hence, it has been difficult to arrive at a useful univalued appreciation of a coal's merit for iron-making.

The study presents a revised theoretical treatment of a univalued measure (net carbon) of the chemical merit of coal, or coke, to iron making. Originally, "effective carbon" was proposed by Flint as an empirical measure of this type. The term "net carbon" was introduced by Callcott to denote such a measure that was derived from a theoretical treatment.

#### BACKGROUND

The total carbon that enters a blast furnace per unit of elemental iron, can be partitioned as in equation (1).

$$CK = NCK + CA + CB + CF + CS \quad (1)$$

where

CK = Carbon content of coke

NCK = Carbon for smelting proper, net carbon in coke

CA = Carbon required for coke ash to be incorporated into  
furnace slag

CB = Carbon used to dissociate moisture in blast

CF = Carbon needed to decompose fluxes

CS = Carbon needed to eliminate coke sulphur

(Units : kg mol/kg mol Fe or %)

Equation (1) can be converted to equation (1a).

$$CK = NCK + CL \quad (1a)$$

where the carbon loss, CL, can be obtained either by empirical methods such as Flint or by theoretical analysis.

Flint was the first to examine the magnitude of the carbon loss terms and, from a linear regression analysis of some blast furnace operating data, obtained simple expressions for the individual terms in equation (1). This approach has been widely accepted and applied in the steel industry but the expressions have needed to be revised in line with advances in blast furnace practice. This empirical approach also has the inherent deficiencies of any regression-based method, viz. multicollinearity among the process variables, misspecification of the regression model, and strictly a limited scope for application of the model's predictions to the range of the original data.

In previous work by Callcott and co-workers at the Central Research Laboratories, Broken Hill Proprietary Co. Ltd., semi-empirical methods have been used to obtain an explicit expression for the carbon loss. In more recent work Cripps-Clark and others have developed a sophisticated thermochemical model for prediction of the effects of changes in burden

and other operating conditions on the performance of specific furnaces. Callcott derived a simple "net carbon" in coal formula and has shown its usefulness in the evaluation of coals.

A purpose of the study is to show how a simple, but conceptually correct thermochemical model of the blast furnace provides an expression for carbon loss which, through equation (1), enables the net carbon in coal to be calculated from the most frequently quoted measures of a coking coal's quality; its ash, sulphur and volatile matter contents.

#### THE CARBON LOSS EXPRESSION

The thermochemical model of the blast furnace is that described by Peacey and Davenport. This model is based upon the pioneering analysis of Rist. The furnace is considered as divided into two segments by its chemical and thermal reserve zones at a temperature of 1200 K, as illustrated in Figure 2.

Coke passes unaltered chemically through the top zone because carbon is not gasified in or above the chemical reserve zone. Coke gasification and combustion takes place in the lower segment so that highly reducing gases pass the chemical reserve zone to the upper segment. Higher iron oxides are reduced by reducing gases to wustite  $Fe_{0.947}O$  by the time they descend into the chemical reserve zone. Reduction of wustite and melting to carbon-rich liquid iron takes place below the chemical reserve zone.

Using the symbols and terminology of Peacey and Davenport all quantities are expressed as  $kgmol/kgmol$  Fe or  $kJ/kgmol$  Fe. The stoichiometric oxygen balance for the bottom segment of the furnace is:

$$n_{OB} + 1.06 + zn_I + 2n_{CaCO_3} = 1.3n_{AC} + 0.38 y n_I \quad (2)$$

The stoichiometric carbon balance for the furnace overall is:

$$n_{AC} + (C/Fe)_m = n_{CC} + x n_I + n_{CaCO_3} \quad (3)$$

The enthalpy balance for the bottom segment of the furnace is:

$$D_{wrz} = n_I D_I = 198000 n_{AC} + E_B n_{OB} + 95000 y n_I \quad (4)$$

where  $D_{wrz}$  is the enthalpy demand for the wustite reduction zone, and  $E_B$  is the enthalpy of the air blast.

#### Incremental Effects of Coke Impurities

**Ash** Consider an incremental increase  $\Delta wt_{ASH}$ , in the amount of ash in the coke which is charged to the blast furnace. This increment will require :-

- an incremental increase in the amount of limestone charged  $\Delta n_{CaCO_3}$ ;
- an incremental increase in the amount of slag formed  $\Delta wt_{slag}$ ;
- an incremental increase in the amount of active carbon used in the bottom segment of the furnace to provide heat to decompose the limestone and smelt the ash  $\Delta n_{AC}$ ;
- an incremental increase in the amount of oxygen in the blast air needed to react with the extra carbon  $\Delta n_{OB}$ ;
- an incremental increase in the amount of water vapour (blast humidity) which enters as an "injectant" at the tuyeres  $\Delta n_I$  ( $x=0$   $y=1$   $z=1$ );  
and thereby an incremental increase in the amount of carbon which must be charged to the furnace as coke,  $\Delta n_{CC}$ .

These incremental changes are related through the mass and enthalpy balance equations (2), (3) and (4) :-

$$\Delta n_{OB} + 2\Delta n_{CaCO_3} = 1.3 \Delta n_{AC} + (0.38 y - z) \Delta n_I \quad (2a)$$

$$\Delta n_{AC} = \Delta n_{CC} + \Delta n_{CaCO_3} \quad (3a)$$

$$\begin{aligned} D_{slag} \Delta wt_{slag} + D_{CaCO_3} \Delta n_{CaCO_3} \\ = 198000 \Delta n_{AC} + E_B \Delta n_{OB} + (95000 y - D_I) \Delta n_I \end{aligned} \quad (4a)$$

Now assuming a typical value for blast humidity of 23 g/Nm<sup>3</sup> (10 gr/scf) it follows that

$$\Delta n_I = 0.069 \Delta n_{OB}$$

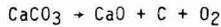
A typical blast temperature of 1400K is also assumed. Thus

$$E_B = 17000 \text{ kJ/kg mole O}$$

and the heat demand of the water vapour "injection" with the blast air is

$$D_I = 240500 \text{ kJ/kg mole H}_2\text{O}$$

Heat demand for decomposition of limestone is given by the enthalpy change of the reaction



$$D_{CaCO_3} = 561000 \text{ kJ/kg mole CaCO}_3$$

Heat demand per kilogram of slag can take the representative value of +250 kJ per kg of slag.

Finally, the incremental increase in the amount of slag is related to the increment in CaCO<sub>3</sub> charged:

$$\Delta wt_{slag} = 56 \left( 1 + \frac{1}{\phi_S - \phi_A} \right) \Delta n_{CaCO_3}$$

where  $\phi_S$  and  $\phi_A$  are the basicity ratios for the slag and coke ash respectively. This relationship assumes that limestone is the only flux added to smelt the coke ash. In this analysis, the addition of a siliceous flux to adjust the  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio in the slag is ignored.

Substitution in the mass and enthalpy balance equations gives :-

$$\Delta n_{\text{CC}} = (0.06772 (1 + \frac{1}{\phi_S - \phi_A}) + 1.7787) \Delta n_{\text{CaCO}_3} \quad (5)$$

Equation (5) relates the amount of extra limestone  $\Delta n_{\text{CaCO}_3}$  that is required to flux an incremental increase in coke ash  $\Delta \text{wt}_{\text{ASH}}$  to the amount of extra coke carbon needed to meet the increased heat demand in the bottom segment of the furnace.

Now

$$\Delta n_{\text{CaCO}_3} = \frac{1}{56} \frac{\phi_S - \phi_A}{\phi_S + 1} \Delta \text{wt}_{\text{ASH}}$$

Thus, by substitution in equation (5) we have an explicit relation between the extra coke carbon needed for an increase in the amount of coke ash :

$$\Delta \text{wt}_{\text{CC}} = \frac{12}{56} [1.7787 + 0.06772 (1 + \frac{1}{\phi_S - \phi_A})] \frac{\phi_S - \phi_A}{\phi_A + 1} \Delta \text{wt}_{\text{ASH}} \quad (6)$$

For coke ash of Australian coals, the effective basicity ratio

$$A = \frac{\text{CaO} + \text{MgO}}{\text{SiO}_2 + \text{Al}_2\text{O}_3} \quad \text{is in the range } 0.01 - 0.05. \quad \text{For modern}$$

blast furnaces the slag basicity ratio  $\phi_S = 1.2$  approximately. For the remainder of this analysis  $\phi_S = 1.2$  and  $\phi_A = 0.025$  are assumed. Thus equation (6) becomes

$$\Delta \text{wt}_{\text{CC}} = 0.468 \Delta \text{wt}_{\text{ASH}} \quad (7)$$

**Sulphur** An incremental increase in sulphur added to the furnace  $\Delta \text{wt}_S$  requires additional limestone to increase the sulphur capacity of the slag. Based on empirical results Henderson found that

$$\Delta \text{wt}_{\text{CaCO}_3} = 30 \Delta \text{wt}_S$$

This additional limestone requires additional coke carbon for decomposition of the limestone, slag formation etc., which may be calculated from equation (5), thus

$$\Delta \text{wt}_{\text{CC}} = \frac{30 \times 12}{100} \left[ 1.7787 + 0.06772 \left( 1 + \frac{1}{\phi_S - \phi_A} \right) \right] \Delta \text{wt}_S \quad (8)$$

For typical basicity ratio values of  $\phi_S = 1.2$ ,  $\phi_A = 0.025$ , this becomes

$$\Delta \text{wt}_{\text{CC}} = 6.855 \Delta \text{wt}_S \quad (9)$$

This analysis ignores any external desulphurisation practice as a second order effect.

#### NET CARBON FORMULAE

##### **Coke**

The foregoing theoretical analysis allows the carbon loss terms in equation (1) to be expressed as follows:

from equation (7) :  $CA + CB + CF = 0.468 \text{ ASH}\%$

from equation (9) :  $CS = 6.855 \text{ S}\%$

Carbon content of the coke can be expressed, after Callcott, as

$$CK = 97 - 0.97 \text{ ASH\%} \quad (10)$$

Thus, net carbon in coke can be expressed by substituting in equation (1):

$$NCK = 97 - 1.438 \text{ ASH\%} - 6.855 \text{ S\%} \quad (11)$$

### Coal

The objective is to express net carbon of coal in terms of coal analyses. This requires appropriate expressions for coke yield %, ash% of coke, and total sulphur of coke.

The net carbon in a coking coal can be treated as a conserved quantity and be defined by the relationship :

$$NCC = \frac{\text{Coke Yield \%}}{100} \cdot NCK$$

After Callcott, coke yield can be expressed :

$$\text{Yield \%} = 96.1 - 0.75 \text{ vm} \quad (13)$$

Ash can be treated as a conserved quantity in carbonisation. Thus,

$$\frac{\text{Ash \% coal}}{\text{Ash \% coke}} = \frac{\text{Yield \%}}{100} \quad (14)$$

Sulphur is not a conserved quantity during carbonisation. The relationship between coke and coal sulphur, proposed by Blayden and Mott is :

$$\text{S\% in coke} = 0.82 \text{ s\% in coal} \quad (15)$$

Substitution of equations (11), (13), (14), and (15) in equation (12) gives :

$$NCC = 93.22 - 0.728 \text{ vm} - 1.438 \text{ ash\%} - 5.40\text{s\%} + 0.0422 \text{ vm.s\%} \quad (16)$$

It is apparent from the foregoing that net carbon in coal can be used as a measure of the chemical merit of a coking coal if it can be assumed that, in any particular ironworks, coals are selected for the blend so that the proportion of the coke make ("wharf coke") which is charged to the blast furnace as lumps ("skip coke") is a constant high figure. This condition is posited on the basis that coke fines, although used in sinter, are of less value than lump coke. It is also necessary to assume that only marginal adjustments are made to a coking blend's proportions and that the marginal change in coke rate due to a marginal change in net carbon of the blend has no effect on net operating costs of the blast furnace (excluding coke costs). In effect, operations are considered to use the cheapest net carbon subject to meeting coke physical quality specifications.

#### NET CARBON AS AN EXPLANATORY VARIABLE FOR COKING COAL PRICES

Callcott evaluated a net carbon model of Japanese coal prices in 1966 in terms of net carbon and of the independent parameters (VM %, ash %,) separately. His study found net carbon to be linearly correlated with price FOB and caking indices to have non-significant linear regression coefficients.

The prices of coking coals imported by Japan have been the subject of later linear regression analysis by Pearson, Kittredge and Sivertson, and Miyazu et al. None of their price models included net carbon of coal as an explicit parameter of the model, although all models included one or more of the variables which determine net carbon; ash %, sulphur % and volatile matter %. The natures of their models are summarised in Table 1.

Essentially, their models have been constructed with a "black-box" attitude to the cokemaking/ironmaking processes, that approach being to examine the statistical significance of every conceivable determining parameter. Only those parameters which were statistically significant were included in the models. The only parameters which the models have in common are the mean maximum reflectance of vitrinite  $\bar{R}_O$  max and ash %, although several of the variables not in common correlate with each other.

A price model which is based upon an analysis of the costs incurred in cokemaking and iron making processes and of the market value of the products of these processes should be more accurate. However such a model is based upon information proprietary to the steelmakers and very little information appears in the literature. Outlines of break-even price models have been given by Brown and Bennett.

Here we examine the hypothesis that net carbon NCC defined by equation (16), is effectively a primary determinant of coal price. From the foregoing theoretical analysis it can be appreciated that net carbon in coal reflects the relative value of a coal with respect to the thermal and chemical requirements of the blast furnace process. It does not account either for the value, if any, of by-products produced from the volatile matter of the coal or for the contribution of a coal to the strength of the coke, and therefore will not be a complete descriptor of the merit of a coking coal.

Caking properties, which influence coke strength, should not affect the penalties accruing to ironmaking costs as a result of ash and sulphur in the coal. Penalties for increasing ash and sulphur should be in the ratio of their coefficients in the net carbon formula, equation (16).

### Coking Coal Price Model

A price model in terms of the contract coal quality specifications could be in form:

$$\text{Price} = \alpha_0 + \alpha_1 \text{NCC} + \alpha_2 \text{TM}' + \alpha_3 \text{CP}$$

where NCC = net carbon in coal, % db

TM' = as shipped total moisture\*, % db

CP = caking property, such as crucible swelling number

$$*\text{TM}' = \frac{100 \text{ TM}}{100 - \text{TM}} \text{ where TM} = \text{As shipped Total Moisture, \% wet coal}$$

The coefficients  $\alpha_1$  and  $\alpha_3$  are expected to be positive and  $\alpha_2$  negative.

Net carbon (dry basis), Specification total moisture (dry basis) and caking property (CSN) for some Australian U.S. and Canadian coking coals are detailed in Table's 2 and 3.

### CONCLUSION

It has been shown how a rigorous thermochemical analysis of the blast furnace and a minimum of assumptions as to slag and ash composition, allows the derivation of a simple formula for the net carbon in coal. Net carbon can be calculated from the results of an analysis for ash, sulphur and volatile matter contents of the coal using the equation (16).

The reader may wish to alter some of the simplifying assumptions in order to achieve closer correspondence with his application. Net carbon is proposed as a convenient measure of the chemical merit of a coking coal which should be useful provided cognisance of the basis of the formula is maintained.

Net carbon appears to be a significant explanatory variable of the price of a coking coal. Together with a caking index such as crucible swelling number and total moisture, net carbon explains some of the variation in prices between coals over recent periods. This report has not set out to provide a comprehensive study of coal market price formation and the price model presented here is not claimed to be definitive but rather is presented to illustrate the usefulness of the net carbon concept.

NOMENCLATURE

ash %	% ash in the coal, dry basis (d.b.)
ASH %	% ash in the coke, d.b.
CA	% carbon in coke to melt ash, d.b.
CB	% carbon in coke to dissociate moisture in the blast, d.b.
CK	% carbon content of coke, d.b.
CF	% carbon in coke to decompose limestone flux requirement of coke ash, d.b.
CS	% carbon in coke to eliminate sulphur in coke, d.b.
NCK	% net carbon in coke, d.b.
NCC	% net carbon in coal, d.b.
s %	% sulphur in coal, d.b.
S %	% sulphur in coke, d.b.
vm%	% volatile matter in coal, d.b.
A	coke ash basicity ratio, $\frac{\text{CaO} + \text{MgO} \%}{\text{SiO}_2 + \text{Al}_2\text{O}_3 \%}$
S	slag basicity ratio
$D_{\text{CaCO}_3}$	enthalpy demand for decomposing charged $\text{CaCO}_3$ to $\text{CaO}$ , $\text{C}$ and $\text{O}_2$ at the thermal reserve temperature (extra heat demand due to charging $\text{CaCO}_3$ rather than $\text{CaO}$ ) kJ (kg mole of $\text{CaCO}_3$ )-1
DI	enthalpy demand for decomposing 1 mole of tuyere injectant at its injection temperature to form its component elements at the thermal reserve temperature kJ (kg mole of injectant)-1
$D_{\text{slag}}$	enthalpy demand for heating slag oxides and forming 1 kg of liquid slag. kJ (kg of slag)-1
Dwrz	total heat demand for wustite reduction zone kJ mole of product $\text{Fe}$ )-1

**NOMENCLATURE contd.**

$E_B$	blast enthalpy	$\text{kJ (kg mole of O)}^{-1}$
$C$	mole ratios in the product hot metal	
Fe		$\text{kg mole (kg mole Fe)}^{-1}$
$n_{AC}$	active carbon taking part in heting or reduction reactions	$\text{kg mole C (kg mole Fe)}^{-1}$
$n_{CC}$	input carbon as coke	$\text{kg mole C (kg mole Fe)}^{-1}$
$n_{CaCO_3}$	charged $\text{CaCO}_3$ kg mole $\text{CaCO}_3$	$(\text{kg mole Fe})^{-1}$
$n_{OB}$	oxygen supplied in blast air	$\text{kg mole O (kg mole Fe)}^{-1}$
$n_I$	moles of tuyere injectant per mole of product Fe	$\text{kg mole (kg mole Fe)}^{-1}$
$x, y, z$	stoichiometry of the injectant, represented by $C_x(H_2)_yO_z$	
$w_{slag}^t$	mass of slag produced	$\text{kg (kg mole Fe)}^{-1}$
$w_{CC}^t$	mass of carbon as coke	$\text{kg (kg mole Fe)}^{-1}$
$w_{ASH}^t$	mass of coke ash charged with coke	$\text{kg (kg mole Fe)}^{-1}$
TM	% total moisture in as-shipped coal, moist basis	
TM'	% total moisture in as-shipped coal, dry basis	
CSN	crucible swelling number	
$\bar{R}_O^{\max}$	mean maximum reflectance of vitrinite	
M.F.	maximum fluidity	
$R^2$	coefficient of multiple determination, a measure of the proportion of the total variation in the dependent variable explained by the regression model.	
S.E.E.	standard error of estimate, the square root of the estimate of the variance of the dependent variable, a measure of how far a given observation on the dependent variable could be from the predicted value.	

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FIGURE 1 - SCHEMATIC OF THE COKE-OVEN/BLAST FURNACE IRONMAKING PROCESS

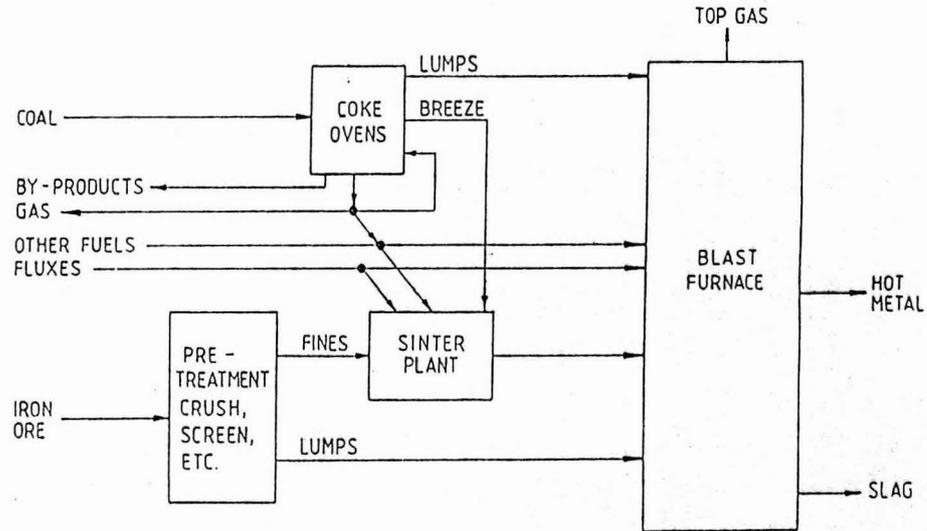
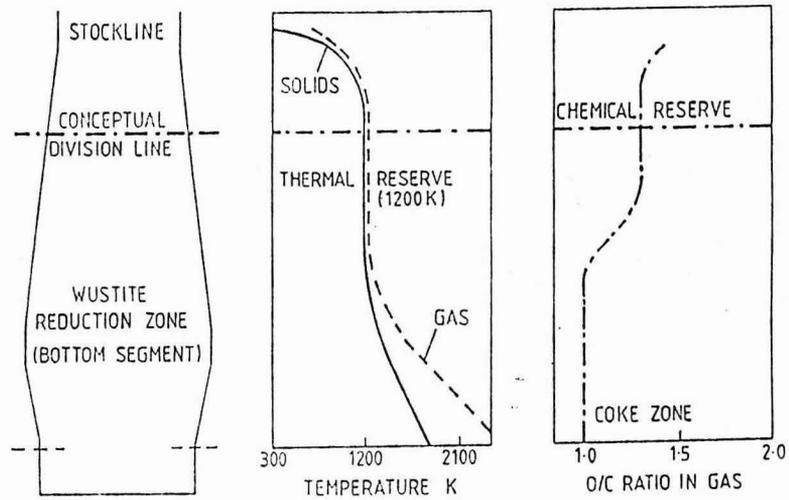


FIGURE 2 - CONCEPTUAL DIVISION OF THE BLAST FURNACE INTO UPPER AND LOWER SEGMENTS



EVALUATION AND SELECTION OF COKING COALS

TABLE 1  
PRICE MODELS OF COKING COALS IMPORTED TO JAPAN

	Callcott	Pearson	Kittredge and Sivertson	Miyazu et al
Year of Data	1966	1978	1977	1979
Source of Coals	Australia USA	USA Canada Australia South Africa	USA Canada USSR Australia South Africa	Not specified
Statistically Significant Parameters ( $\alpha = .05$ )	Net Carbon or ash % vm %	CSN $R_0$ max ash %	$\bar{R}_0$ max CSN M.F. ash % s % TM % Factors for: Transport Productivity Contract Term	$\bar{R}_0$ max Reactives % Organic Inerts % log (M.F.) ash % s %
No of Coals	57	30	36	51
S.E.E.	-	4.4	3.26	-
R <sup>2</sup>	-	.80	.91	.856

TABLE 2 - AUSTRALIAN COKING COALS

COAL BRAND	TOTAL MOISTURE %	PROXIMATE ANALYSIS (A.D BASIS)			TOTAL SULPHUR (A.D) %	FSI	NET CARBON (D.B.)	TOTAL MOISTURE (D.B.)
		MOISTURE %	ASH %	VOLEATILE MATTER %			%	%
NORWICH PARK	8.0	0.9	9.5	17.2	0.65	8-9	63.71	8.70
SARAJI	8.0	1.0	9.3	19.5	0.55	8-9	62.80	8.70
PEAK DOMES	8.0	1.0	9.3	21.0	0.55	8-9	61.74	8.70
GERMAN CREEK	8.0	1.2	8.5	21.0	0.70	8-9	62.18	8.70
RIVERSIDE	8.0	1.4	9.8	23.9	0.57	7.5	58.74	8.70
GOONYELLA	8.0	1.0	8.0	25.5	0.50	8	60.65	8.70
OAKY CREEK	8.0	1.2	8.0	29.5	0.80	8-9	56.48	8.70
GREGORY	8.0	2.0	8.0	32.0	0.65	8-9	55.06	8.70
CURRAGH	8.0	1.5	7.0	22.0	0.52	7	64.37	8.70
BLACKWATER	8.0	2.0	7.8	27.0	0.50	6	59.50	8.70
MURA	8.0	2.0	8.0	28.0	0.45	8	58.76	8.70
WESTCLIFF	7.0	1.0	9.8	21.5	0.38	5-7	61.47	7.53
TAMBOOR	7.0	1.2	8.5	27.5	0.40	6	58.90	7.53
MADQUARIE	8.0	2.5	7.5	34.5	0.45	6-7	54.61	8.70
HEWDELL	7.0	3.2	8.5	37.5	0.60	5	50.06	7.53

TABLE 3 - CANADIAN, U.S. AND POLISH COKING COALS

COAL BRAND	TOTAL MOISTURE %	PROXIMATE ANALYSIS (A.D BASIS)			TOTAL SULPHUR (A.D) %	FSI	NET CARBON (D.B.)	TOTAL MOISTURE (D.B.)
		MOISTURE %	ASH %	VOLATILE MATTER %			%	%
SMOKY RIVER	6.0	1.0	7.0	18.0	0.50	7-9	67.40	6.38
BALMER	8.0	1.0	9.5	22.0	0.40	6-8	61.47	8.70
FORDING	8.0	1.0	9.5	22.5	0.45	5-7	60.89	8.70
FORDING H.V.	8.0	1.5	6.5	30.0	0.60	6-8	59.02	8.70
DEVCO	5.0	2.0	3.0	35.5	1.20	6-8	57.68	5.26
BEATRICE	5.0	1.0	4.8	18.0	0.70	8-9	69.78	5.26
NEW RIVER	5.0	1.0	5.7	18.0	0.75	8-9	68.11	5.26
ROBINSON PHILLIPS	6.0	1.0	6.2	24.5	0.75	8-9	62.87	6.38
PITTSTON M.V.	6.0	1.0	5.5	29.5	0.75	8-9	60.32	6.38
PEERLESS	6.0	2.0	6.3	32.0	0.75	7-9	57.12	6.38
COAL MOUNTAIN	5.0	2.0	6.0	33.0	0.90	7-9	56.25	5.26
FIRST MAJA	8.0	2.0	7.3	28.5	0.67	8	58.56	8.70

