

The estimation of some properties of South African coals from proximate analyses

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SYNOPSIS

Correlations have been obtained whereby calorific values, ultimate analyses, and Hardgrove Indices can be estimated from proximate analyses of South African coals. This has been done by the application of stepwise multiple linear-regression analyses of a mass of data published by the Fuel Research Institute.

SAMEVATTING

Korrelasies is verkry deur middel waarvan kaloriewaardes, elementanalises en Hardgrove indekse geskat kan word van kortanalises vir Suid-Afrikaanse steenkole. Dit is gedoen deur die toepassing van trapsgewyse veelvoudige lineêre regressie analises op 'n massa van gegewens wat deur die Brandstofnavorsingsinstituut gepubliseer is.

INTRODUCTION

To contend with the wide range of uses to which coal is put and the highly variable nature of this commodity, a multitude of various tests have been developed for the characterization of coal. The simplest and most popular of these is the proximate analysis, which comprises determinations of inherent moisture, ash, and volatile matter. Fixed carbon is found by difference. Calorific value, sulphur content, ash fusion temperature, and swelling index are often reported together with the proximate analysis. Few tests other than these are done on a routine basis by quality-control laboratories, as they tend to be tedious and expensive to perform.

With the growing sophistication of consumers, however, more extensive specifications are being made, entailing a considerable increase in analytical work. It would be very useful indeed to have methods available whereby coal properties can be estimated from a number of simple tests.

As a first step in this direction, this paper deals with an adaptation to South African conditions of a study reported some years ago by Humphreys and Lawrence¹.

In contrast to the classical work of Seyler², whereby some properties are predicted from ultimate analyses, Humphreys and Lawrence showed that the ultimate analysis, calorific

value, and Hardgrove Index can be estimated from the proximate analysis with reasonable accuracy. This has also been found to be true for South African coals, particularly for the first two factors.

TYPES OF CORRELATIONS

The standard expression used in multiple linear regression analysis is:

$$Y = a_0 + a_1 X_1 + a_2 X_2 + a_3 X_3 + \dots \quad (1)$$

For this study, the dependent variable Y may be the calorific value, Hardgrove Index, total carbon, etc. The independent variables X_1, X_2, X_3 , etc. may be percentage moisture (H_2O), percentage volatile matter (VM), or percentage ash (Ash) on their own, but also powers, functions, or any combination of these.

Humphreys and Lawrence had considerable success using the simplest polynomial form, and this will be adhered to. That this form is reasonable, at least for calorific value, can be shown by the following argument.

If, as a first approximation, the calorific values of volatile matter and fixed carbon are assumed to be

constant irrespective of rank and equal to α and β respectively, then the calorific value (CV) of any coal on a dry, ash-free (d.a.f.) basis is:

$$CV = \alpha \frac{(VM)}{100} + \beta \frac{(100 - VM)}{100}$$

Volatile matter and fixed carbon are expressed on a d.a.f. basis, i.e., fixed carbon = $(100 - VM)$. The above expression can be rearranged to give

$$CV = \beta + (a - \beta) \frac{(VM)}{100}$$

or

$$CV = a_0 + a_1 (VM) \dots \dots (2)$$

If the calorific values of volatile matter and fixed carbon are linearly dependent on the rank of the coal and moisture is taken as an indication of rank, then on the same basis as before:

$$CV = a_0 (1 + a_1 (H_2O)) \frac{(VM)}{100} + \beta_0 (1 + \beta_1 (H_2O)) \frac{(100 - VM)}{100}$$

which reduces to the form

$$CV = a_0 + a_1 (VM) + a_2 (H_2O) + a_3 (VM) (H_2O) \dots \dots (3)$$

If these variables are expressed on an air-dry basis, rather than d.a.f., equation (3) becomes

$$\frac{CV}{\left(1 - \frac{Ash + H_2O}{100}\right)} = a_0 + a_1 \frac{VM}{\left(1 - \frac{Ash + H_2O}{100}\right)} + a_2 (H_2O) + a_3 (H_2O) \frac{VM}{\left(1 - \frac{Ash + H_2O}{100}\right)}$$

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which, on rearrangement, gives

$$CV = a_0 + a_1 (VM) + a_1 (H_2O) + a_3 (H_2O) (VM) + a_4 (H_2O) (Ash) + a_5 (H_2O)^2 + a_6 (Ash) \dots \dots \dots (4)$$

This line of argument can be extended to allow for heat liberated during the ashing of mineral matter, and so on, and similar expressions will be obtained. In this work the most convenient and accurate expressions of this general type will be found empirically without any regard to the theoretical significance of the various terms.

METHOD OF CORRELATION

The process of correlating a single dependent variable against one or more independent variables using the form of equation (1) is a well-known statistical method, namely multiple linear regression analysis.

A standard method for such a regression is the Forward Selection Procedure. By this method, variables are inserted into the regression equation in turn, the order of insertion being determined by the use of the partial correlation coefficient as a measure of the importance of variables not yet in the equation. A statistical significance test is used, and the process is terminated when the contribution of the most recently entered variable is non-significant.

The Stepwise Procedure is an improved version of the Forward Selection Method. The improvements involve the re-examination, at every stage of the regression, of the variables incorporated into the model in previous stages. A variable that may have been the best single variable to enter at an early stage may at a later stage be superfluous, because of the relationships between it and the other variables now in the regression. To check on this, the significance of each variable in the regression at any stage of calculation is evaluated. This provides a judgement on the contribution made by each variable as though it had been the most recent variable entered, irrespective of its actual point of entry into the model. Any variable that provides a non-significant contribution is removed from the model. The process is continued until no more variables will be admitted to

the equation and no more are rejected.

A UNIVAC subroutine — RES-TEM — was used for this analysis.

CORRELATION OF CALORIFIC VALUE

The data for this correlation were taken from the forty-first Annual Report of the Fuel Research Board (1971). The moisture, ash, and volatile-matter contents of about 250 different coals are listed, together with their calorific values, in MJ/kg. All values are on an air-dry basis.

Humphreys and Lawrence did not attempt to correlate calorific value directly with the proximate analysis. Instead, their method entails the estimation of calorific value from ultimate carbon, which in turn is estimated from volatile matter (d.a.f.). This is cumbersome, and the logical first step is to attempt to correlate calorific value directly with volatile matter (d.a.f.). Using the form of equation (2), the following correlation was found:

$$CV = 36,131 - 0,089 VM \text{ MJ/kg.} \dots \dots \dots (5)$$

This prediction equation has a standard error of 1,0 MJ/kg, which corresponds to roughly 3 per cent, since most calorific values (d.a.f.) are in the range 30 to 35 MJ/kg.

To improve the accuracy of the prediction, equation (5) was extended to include the other proximate analyses, with the following result (d.a.f. basis):

$$CV = 37,454 - 0,032 VM - 0,847 H_2O - 0,051 Ash \dots \dots \dots (6)$$

An interesting feature of this correlation is that the variable that is singly most highly correlated with calorific value is the moisture content — the correlation coefficient is 0,91.

Since moisture, volatiles, and ash were all significant in the prediction of calorific value, an attempt was made to improve the correlation by consideration of their second-order and interaction terms as well. This resulted in the regression equation

$$CV = 37,360 - 1,600 H_2O + 0,055 Ash + 0,231 H_2O^2 - 0,0016 (VM) (Ash) - 0,040 (H_2O) (Ash) \dots \dots \dots (7)$$

This equation has a standard error of 0,35 MJ/kg and is the most accurate prediction equation obtained. However, it is rather unwieldy and would certainly not be easy to represent graphically.

Several other correlations were tried, and are summarized in Table I.

In the development of the various prediction equations, the insertion of more and more terms for the sake of small reductions in the standard deviation was avoided. Equations of reasonable accuracy were sought that could be represented graphically if possible.

The first correlation (equation 5), which involves only volatile matter, has a standard error of 1,00 MJ/kg, or 3 per cent. This is very easy to plot—just one straight line—but the prediction is not very accurate.

The second correlation (equation 6) involves first-order terms of volatiles, ash, and moisture, and has a standard error of 0,44 MJ/kg or 1,5 per cent. This can be fairly easily represented as two straight-line graphs.

The third correlation (equation 7) is more complicated to plot. Second-order and interaction terms are involved, necessitating at least three graphs. Furthermore, these graphs would be difficult to read, since interpolation would not be linear.

Thus, although equation (6) is statistically less accurate than equation (7), it is, from the practical viewpoint, a better correlation. For the same reason, this equation is also better than the correlations of Table I.

Equation (6) is represented graphically in Fig. 1. This graph permits more than 83 per cent of the values to be predicted within 0,6 MJ/kg, or 2 per cent. Over 97 per cent of the predicted values have an error of less than 0,9 MJ/kg, or 3 per cent. This is quite satisfactory, and it is therefore suggested that, for quick predictions of calorific value, the graph of Fig. 1 should be used.

Table II shows calorific values obtained from Fig. 1, together with the actual values, for several random points.

TABLE I
SUMMARY OF CORRELATIONS OF CALORIFIC VALUE

Variables considered	Resulting equation	Std. error MJ/kg	Comments
VM, H ₂ O, Ash VM ² , H ₂ O ² , Ash ² , (VM) (H ₂ O), (VM) (Ash), (H ₂ O) (Ash)	CV = 37,927 - 0,076VM - 1,830H ₂ O - 0,338Ash + 0,0014VM ² + 0,203H ₂ O ² - 0,027 (H ₂ O) (Ash)	0,28	All values on air-dry basis. Average CV's about 27MJ/kg
FC, FC ² , FC ³ , (FC/VM), (FC/VM) ² , (FC/VM) ³	CV = - 3,793 + 0,812FC - 6,212 (FC/VM) - 0,00002 (FC) ³ + 0,710 (FC/VM) ² - 0,028 (FC/VM) ³	0,68	All values on air-dry basis
VM, H ₂ O, Ash VM ² , H ₂ O ² , Ash ² , (VM) (H ₂ O), (VM) (Ash), (H ₂ O) (Ash)	CV = 37,639 - 0,077VM - 1,254H ₂ O - 0,052Ash + 0,0012VM ² + 0,145H ₂ O ² - 0,00027 (VM) (Ash) - 0,025 (Ash) (H ₂ O)	0,35	All values on dry, ash-free basis

CORRELATION OF ULTIMATE ANALYSES

The data for these correlations were taken from a publication⁴ of the Fuel Research Institute of South Africa. For each sample, the volatile-matter content and the percentages of ultimate carbon, hydrogen, and nitrogen are reported on a dry, ash-free basis, and ash is given on an air-dry basis. Moisture analyses were not included, but the appropriate figures were supplied on request by the Fuel Research Institute. The basis of each analysis was maintained in the regression analyses.

Correlations were obtained for carbon, hydrogen, and nitrogen. The variable that singly correlated best with all of these is volatile matter. In all cases, ash was eventually left out of the regression, and a correlation involving only moisture, volatiles, and their second-order terms was sought.

Ultimate Carbon

The first correlation attempted was for ultimate carbon, using VM, H₂O, and Ash. This resulted in

$$\begin{aligned} \%C = & 96,097 - 0,274 \text{ VM} \\ & - 0,978 \text{ H}_2\text{O} - 0,060 \text{ Ash} \end{aligned} \quad \dots \dots \dots (8)$$

with a standard error of 0,71. Since volatiles and moisture entered the regression equation first and are thus the most significant input variables, an expression was sought including only these variables, with their second-order and interaction terms, to attempt to improve the accuracy. This gave the prediction equation

$$\begin{aligned} \%C = & 97,116 - 0,429 \text{ VM} \\ & - 1,110 \text{ H}_2\text{O} + 0,00315 \text{ VM}^2, \end{aligned} \quad \dots \dots \dots (9)$$

which has a standard error of 0,73.

As far as accuracy is concerned,

there is not much to choose between equations (8) and (9). From the practical point of view, however, equation (9) is easier to represent graphically, since equation (8) re-

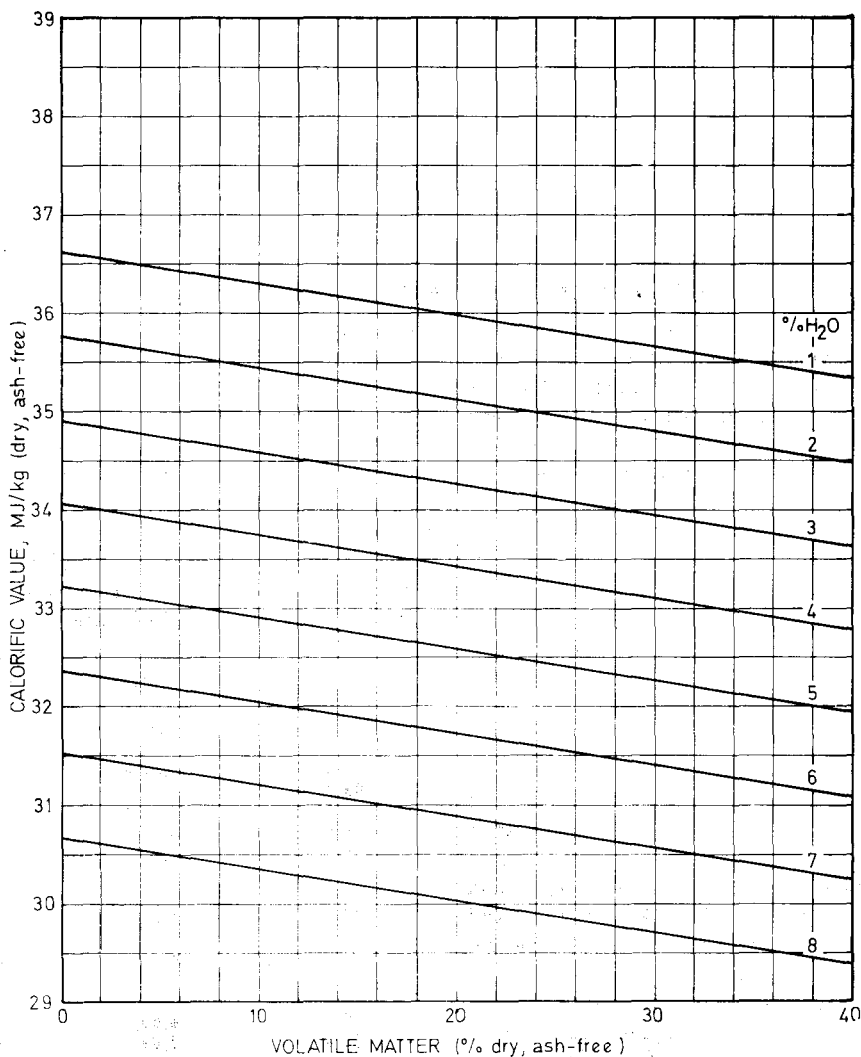


Fig. 1 (a) — The prediction of calorific value

TABLE II
ACTUAL AND PREDICTED CALORIFIC VALUES FOR RANDOM SAMPLES (AIR-DRY BASIS)

VM	H ₂ O	Ash	CV (actual)	CV (predicted)
28,4	2,9	20,0	25,1	25,3
10,4	1,6	11,2	30,7	30,6
23,8	5,2	22,4	21,8	22,3
18,7	1,3	19,6	27,7	27,4
9,7	1,4	7,6	32,4	32,3

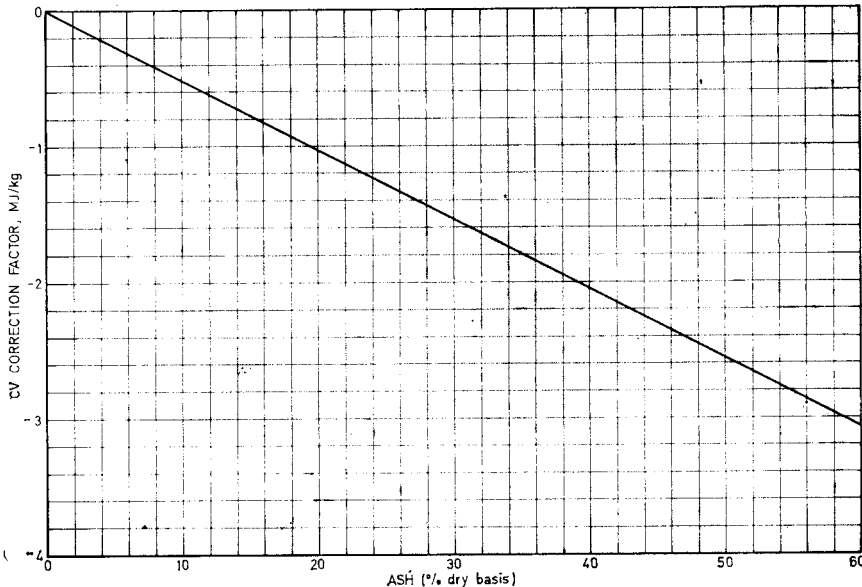


Fig. 1 (b)—The prediction of calorific value. Ash correction

quires at least two graphs.

The graph of equation (9) appears in Fig. 2. From this graph, ultimate carbon is predicted within 1 per cent of its true value for over 85 per cent of the time, and within 2 per cent for over 98 per cent of the time. This graph is thus recommended for the prediction of ultimate carbon.

Ultimate Hydrogen

The equivalent of equation (8) for hydrogen gave the following correlation:

$$\%H = 3,306 + 0,057 VM - 0,157 H_2O \dots (10)$$

with a standard error of 0,18.

The fact that Ash is not entered into the regression indicates that it is not significant in predicting ultimate hydrogen. A new regression was tried, with VM and H₂O and their second-order terms. The resulting equation,

$$\%H = 4,292 + 0,028 VM - 0,675 H_2O + 0,010 (VM)(H_2O) + 0,022 H_2O^2 + 0,00023 VM^2, \dots (11)$$

has a standard error of 0,14.

For over 83 per cent of all coals examined, this last equation predicts ultimate hydrogen percentage within 0,2 of its real value; and over 96 per cent of the predicted values differ from the actual values by less than 0,3.

Equation (11) is difficult to plot graphically. When the last two variables (H₂O² and VM²) are re-

moved, the regression expression becomes

$$\%H = 4,192 + 0,029 VM - 0,593 H_2O + 0,013 (VM)(H_2O) \dots (12)$$

with a standard error of 0,15. This equation is easily represented graphically and appears in Fig. 3. Linear interpolation is possible, and this graph is thus recommended for predictions of ultimate hydrogen.

Ultimate Nitrogen

Nitrogen was the last of the ultimate analyses for which a correlation was attempted. With first-order terms of ash, volatile matter, and moisture content, the equation that results from the computer correlation is

$$\%N = 2,597 - 0,013 VM - 0,029 H_2O \dots (13)$$

with a standard error of 0,124.

Ash, it will be noted, was once again left out of the prediction equation, and second-order terms of only volatiles and moisture were tried. This yielded the equation

$$\%N = 2,965 - 0,032 VM - 0,175 H_2O + 0,017 H_2O^2 + 0,00044 VM^2, \dots (14)$$

which has a standard error of 0,116. From this equation, the predicted values of over 90 per cent of all samples are within 0,2 of their real value.

However, although equation (14) is more accurate than equation (13), the latter expression is far easier to plot (Fig. 4). Thus, for prediction purposes, equation (14) should be used if a calculator or computer is available, but, for rapid estimates, the use of Fig. 4 is recommended.

Other Elements

The correlation coefficient of all

TABLE III
ACTUAL AND PREDICTED ULTIMATE ANALYSES FOR RANDOM SAMPLES

%C	%H	%N	%S	%O
(83,2) 83,2	(4,82) 4,80	(2,03) 2,03	(0,34) (0,34)	9,6 9,6
(86,6) 86,7	(4,45) 4,36	(2,28) 2,20	(1,01) (1,01)	5,6 5,7
(84,1) 85,1	(4,39) 4,51	(1,93) 2,10	(0,36) (0,36)	9,2 7,9
(77,8) 78,1	(4,28) 4,57	(2,00) 1,97	(0,32) (0,32)	15,6 15,0
(82,1) 82,4	(4,80) 5,09	(2,13) 2,10	(0,44) (0,44)	10,5 10,0

Measured values are shown in parentheses

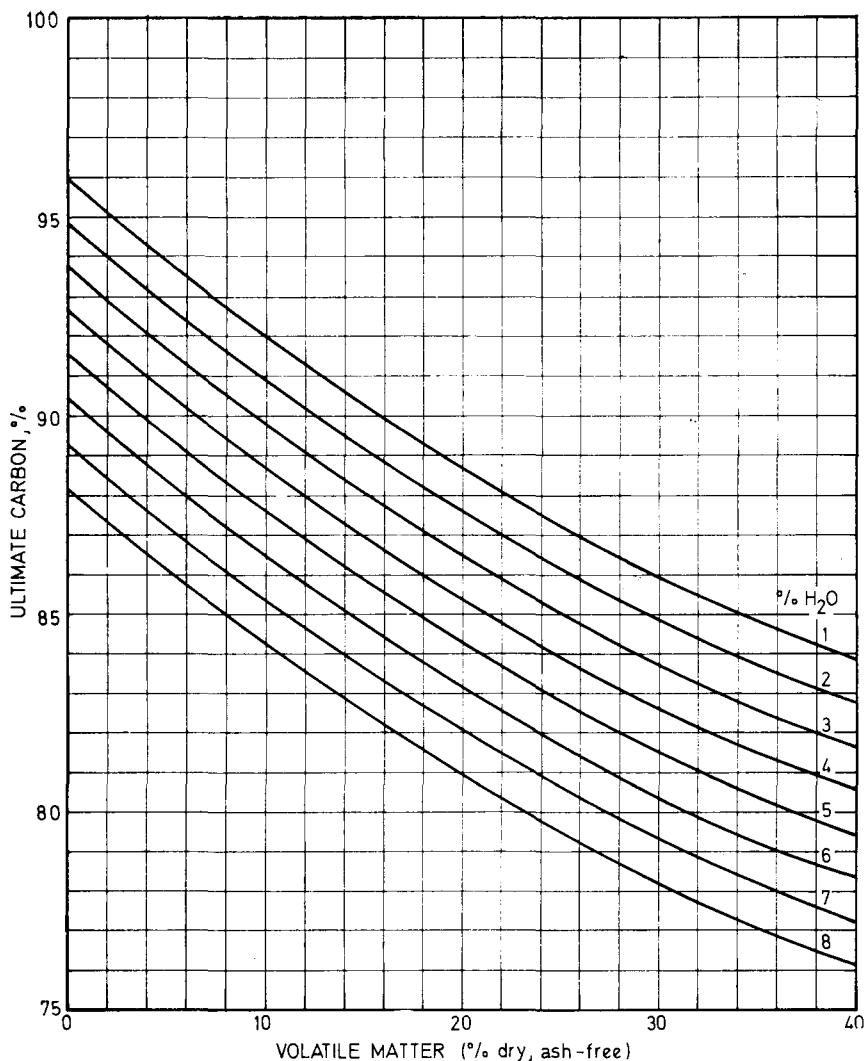


Fig. 2—Prediction of ultimate carbon

other variables with sulphur is very low, and a satisfactory correlation could not be obtained. Sulphur analyses are usually available and are reported together with the proximate analyses. Oxygen is determined by difference and includes all errors of analysis or prediction.

Accuracy

Table III is a comparison of actual and predicted ultimate analyses for several random samples. In each case, the most accurate correlation was employed. As can be seen, the degree of accuracy achieved would be more than adequate for a rapid estimate of ultimate analysis parameters.

CORRELATION OF HARDGROVE GRINDABILITY INDEX

The Hardgrove Grindability Index (HGI) of about 150 samples re-

ported in 1965⁵ and a similar number of more recent results⁶ were correlated with moisture and ash expressed on an air-dry basis and with volatile matter and total sulphur expressed on a dry, ash-free basis.

As experienced by Humphreys and Lawrence, really accurate prediction equations could not be obtained. Their final correlation involved volatile matter, ash, sulphur, and all second-order and interaction terms.

When this form of equation was applied to South African data, the interesting results emerged that ash is of minor importance in the prediction of HGI, and that sulphur hardly correlates with HGI at all. Sulphur content was therefore not considered further in this work.

An examination of the data, and

of plots of HGI against volatile matter^{5, 7} further revealed that there are two distinct bands, one below 15 per cent volatiles, and the other above a volatile-matter content of about 20 per cent. Since most of the data points fall in the latter range, the former band was excluded from some correlations in an effort to improve the accuracy. A summary of the various correlations obtained is given in Table IV.

The most accurate prediction equation is number (17), with a standard error of 5.64. This is, however, not spectacularly better than equation (18) or (19). The inclusion of ash and moisture in the regression therefore makes little improvement in precision. Equation (18) is the most convenient for graphical representation.

To examine whether the inclusion of ash and moisture terms makes any worthwhile improvement to the correlation, a polynomial in powers of volatile matter alone was tried for all the data, and equation (20) resulted. The standard error is a little greater than for the other correlations, despite there being only three terms.

It must be concluded that the Hardgrove Index cannot be inferred satisfactorily from proximate analyses alone. If equation (17) is used, 94 per cent of the predictions will be within 10 points of their actual value, and 82 per cent within 7 points. This is the same order of accuracy achieved by Humphreys and Lawrence, but, as the normal range of HGI values is 30 to 90 points, the percentage error is large.

SUMMARY AND CONCLUSIONS

The calorific value and ultimate analysis of South African coals can be predicted from their proximate analyses with reasonable accuracy. The predictions of the Hardgrove Index are much less satisfactory, but, for lack of other alternatives, various prediction equations that can be used are presented.

Improvements to these correlations could probably be made if they were restricted to coals from various areas, or even to collieries within an area.

It should be possible to extend this work to other cases such as the prediction of the Hardgrove Index from

TABLE IV
SUMMARY OF CORRELATIONS OF HGI

Variables considered	Resulting equation	Std error	Comments
VM, VM ² , VM ³ , H ₂ O, H ₂ O ² , H ₂ O ³ , (VM) (H ₂ O), (VM) ² (H ₂ O) (VM) (H ₂ O) ²	HGI = -29,029 + 0,00558 VM ³ - 0,495VM ² + 13,190VM + 0,123 (VM) (H ₂ O) ² - 1,531 (H ₂ O) ² - 0,188 (H ₂ O) ³ - 0,00849 (H ₂ O) (VM) ²(15)	6,43	All data considered
As above	HGI = 140,185 - 2,595VM + 0,00888 (H ₂ O) (VM) ² - 0,258 (H ₂ O) (VM)(16)	6,36	Data with VM content less than 20% not considered
VM, VM ² , H ₂ O, H ₂ O ² , Ash, Ash ² , S, S ² , (VM) (H ₂ O), (VM) (Ash), (VM) (S), (H ₂ O) (Ash), (H ₂ O) (S), (Ash) (S)	HGI = 184,416 - 5,931VM + 0,208 (H ₂ O) (Ash) + 0,067VM ² - 5,430 (H ₂ O) + 0,301H ₂ O ² - 0,008 (Ash) ²(17)	5,64	Data with VM content less than 20% not considered
VM, VM ² , VM ³ , Ash, Ash ² , Ash ³	HGI = 137,003 - 3,190VM + 0,394Ash + 0,00054VM ³(18)	6,19	Data with VM content less than 20% not considered
VM, VM ² , VM ³ , Ash, Ash ² , Ash ³ , (Ash) (VM), (Ash) (VM) ² , (VM) (Ash) ²	HGI = 164,062 - 4,209VM + 0,00073 (Ash) (VM) ² + 0,0006VM ³ - 0,00826Ash ²(19)	6,09	Data with VM content less than 20% not considered
VM, VM ² , VM ³ , VM ⁴ , VM ⁵ , VM ⁶ , VM ⁷ , VM ⁸ , VM ⁹	HGI = 158,188 - 4,549VM + 0,047VM ²(20)	6,63	Data with VM content less than 20% not considered
As above	HGI = 20,738 + 0,503VM ² - 1,914x10 ⁻² VM ³ + 0,173x10 ⁻⁶ VM ⁶ - 10 ⁻¹² VM ⁹(21)	6,59	All data considered

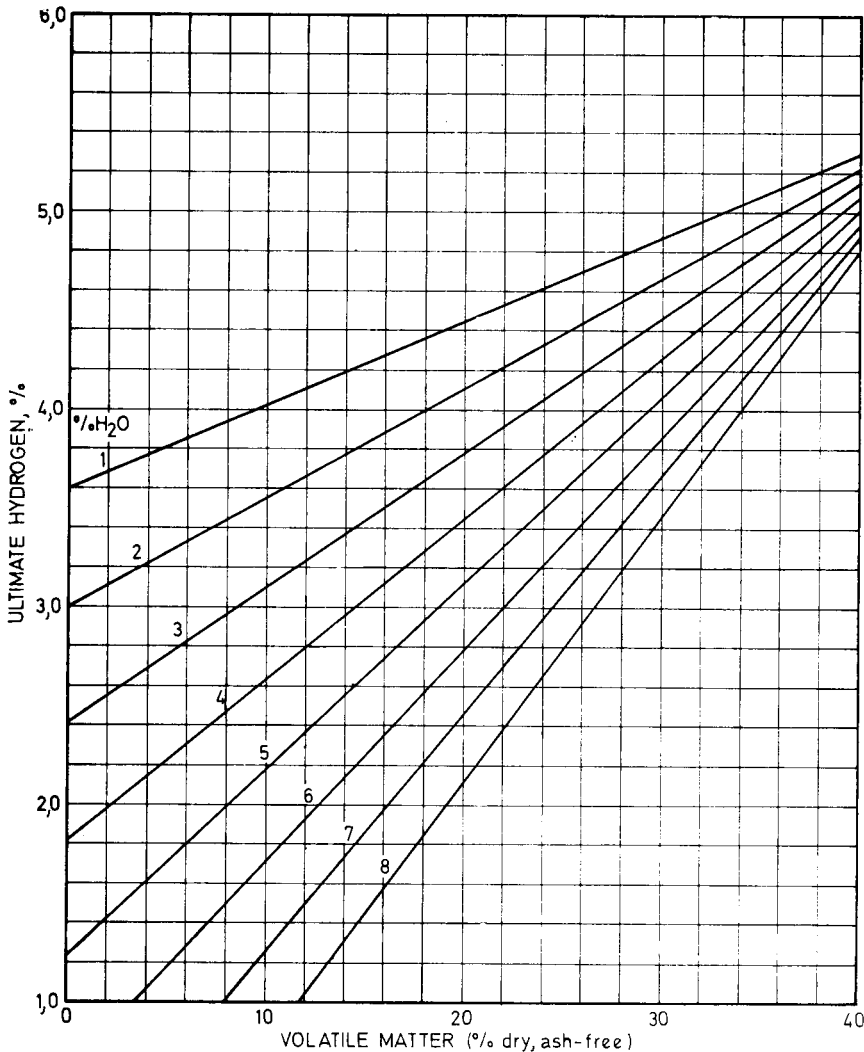


Fig. 3—Prediction of ultimate hydrogen

petrographic and proximate analyses, abrasive index from mineral composition, and coke strength from the various specific laboratory analyses.

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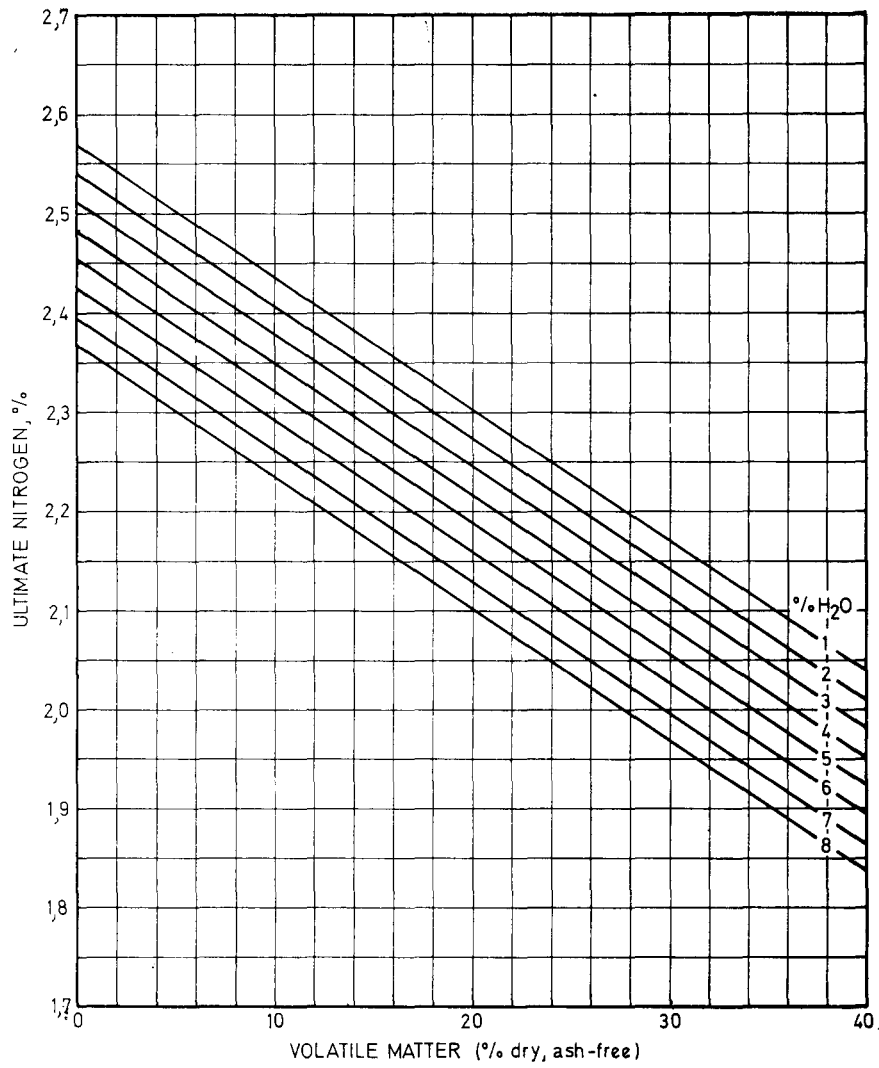


Fig. 4—Prediction of ultimate nitrogen