INDONESIAN MINING JOURNAL Vol. 17, No. 1, February 2014 : 10 - 19

DEVELOPMENT OF NEW EQUATIONS FOR ESTIMATING GROSS CALORIFIC VALUE OF INDONESIAN COALS

PENGEMBANGAN PERSAMAAN BARU UNTUK MENGHITUNG NILAI KALOR BATUBARA INDONESIA

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ABSTRACT

Numerous empirical equations have been published to correlate the gross calorific value (GCV) of coals with the result of proximate or ultimate analysis, however, many researchers continue to propose new equations. One of the reasons is that many existing equations are likely fitted to coal of one region only. This study is aimed to evaluate the applicability of some existing equations to calculate GCV of Indonesian coal and to develop new equations that more accurate to predict the calorific value of Indonesian coal. Ten (10) new GCV formulas based on proximate analysis data of Indonesian coal were generated using SPSS software. They include three (3) equations with one independent variable, four (4) equations with two independent variables, two (2) equations with three independent variables and one (1) equation with four independent variables. The best equation has the following form: GCV= 25.284 (M) + 30.572 (Ash) + 62.127 (VM) + 138.117 (FC) - 2890.095. The result is in agree with previous work that equation involving four independent variables i.e. moisture (M), ash, volatile matter (VM) and fixed carbon (FC) provides the most accurate estimation of GCV. The new equation when it is used for calculating GCV of Indonesian coals gives more accurate results than that of some existing equations in the literatures.

Keywords: gross calorific value, proximate analysis, SPSS, Indonesian coal

SARI

Banyak persamaan empiris telah dibuat yang mengkorelasikan nilai kalor kotor (gross calorific value/GCV) batubara dengan hasil analisis proksimat atau analisis ultimat. Meskipun demikian masih banyak persamaan baru yang terus diusulkan dan salah satu alasannya adalah persamaan-pesamaan yang ada diperkirakan hanya cocok untuk menghitung nilai kalor batubara dari daerah tertentu saja. Penelitian ini bertujuan untuk mengevaluasi beberapa persamaan yang terdapat di literarur dan untuk mengembangkan persamaan baru yang lebih akurat untuk memprediksi nilai kalori batubara Indonesia. Sepuluh (10) persamaan baru yang mengkorelasikan data hasil analisis proksimat batubara Indonesia dengan GCV telah dibuat dengan menggunakan perangkat lunak SPSS. Persamaan yang dikembangkan mencakup tiga (3) persamaan dengan satu variabel bebas, empat (4) persamaan dengan dua variabel bebas, dua (2) persamaan dengan tiga variabel bebas dan satu (1) persamaan memiliki empat variabel bebas. Korelasi terbaik memiliki bentuk sebagai berikut : GCV = 25,284 (M) + 30,572 (Ash) + 62,127 (VM) + 138,117 (FC) - 2.890,095. Hasil ini sejalan dengan hasil penelitian sebelumnya yang menyimpulkan bahwa persamaan yang melibatkan empat variabel bebas yaitu kadar air lembab (moisture/M), abu (ash), zat terbang (volatile matter/VM) dan karbon tertambat (fixed carbon/FC) memberikan estimasi yang paling akurat untuk GCV. Persamaan baru hasil dari kajian ini menghasilkan perkiraan nilai GCV yang lebih akurat dibandingkan persamaan-persamaan lain yang ada di literatur jika digunakan untuk batubara Indonesia.

Kata kunci: nilai kalor kotor, analisis proksimat, SPSS, batubara Indonesia.

Received : 09 October 2013, first revision : 26 November 2013, second revision : 23 January 2014, accepted : February 2014

INTRODUCTION

Since the year 2000, global coal consumption has grown faster than any other fuels. Approximately 27% of the world's primary energy and 41% of the world's electrical energy supply comes from coal. In some countries the percentage of coal-fired power plants is much higher for example in South Africa 93%, Poland 92%, China 79%, and Australia 77% (IEA, 2011). Although growth in world coal consumption experiencing a slowdown in 2012, it is expected the growth to recover in 2014. Growth in coal consumption in 2012 was 2%, much lower than the growth in 2011 which reached 9% (Jones, 2013).

Indonesia is the world's largest thermal coal exporter after overtaken Australia's position in 2011 (www.worldcoal.org). The 5 Largest Indonesian coal export destinations were China, South Korea, India, Japan, and Taiwan (Harrington and Trivett, 2012). Since the use of Indonesian coal is increasing globally, the properties of Indonesian coal should be studied comprehensively.

The calorific value or heat of coal combustion is one of the most important properties of thermal coal since it determines the price of thermal coal and it is usually used as parameters for design calculations of thermal systems. The calorific value of coal can be measured by employing an adiabatic bomb calorimeter or can be calculated from the results of ultimate and proximate analysis using definite equation. The use of equation rather than measure the calorific value experimentally would save cost and time. The equation is also useful for designing calculations and numerical simulations of coal combustion, gasification and pyrolysis. In addition, the equation can be used to evaluate the reliability of the measurements; if a measurement by bomb calorimeter gives ambiguous result, the equation may be used to confirm the result of the measurement.

Numerous empirical equations have been published to correlate the calorific value of fuels with the result of proximate or ultimate analysis. Dulong postulated in the early 1800s that the gross calorific value (GCV) of a sample can be determined from its elemental composition (Buckley and Domalski, 1988). By the year of 1980 at least 9 different formulas for calculating GCV from the ultimate analysis and 11 formulas for calculating GCV from the proximate analysis have been developed (Mason and Gandhi, 1980).

Despite such numbers of equations are available, some researchers (Parikh et al., 2005; Akkaya, 2009; Cordero et al., 2001; Mesroghli et al., 2009) continue to propose new equations. One of the reasons is that the existing equation is likely fitted to coal of one region only (Parikh et al., 2005).

This study is aimed to evaluate the applicability of some existing GCV equations for Indonesian coal and to develop new GCV equations that more accurate to predict the calorific value of Indonesian coal. Although correlations developed from the results of ultimate analysis are considered more accurate, in this study the correlation was developed based on proximate analysis due to limited data available for ultimate analysis. Also, ultimate analysis needs more expensive equipment and highly skilled and dedicated analysts than proximate analysis. The new developed GCV equations will use calorie/gram (adb) instead of BTU/lb or mJ/kg since calorie/gram is more commonly used for expressing calorific value of coals in Indonesia.

LITERATURE REVIEW

Heating value or calorific value of a coal is defined as the amount of heat evolved when a unit weight of coal is burnt completely. It may be reported on two bases, the higher heating value (HHV) or gross calorific value (GCV) and the lower heating value (LHV) or net calorific value (NCV). The GCV refers to the heat released from the fuel combustion with the original and generated water in a condensed state, while the NCV is based on gaseous water as the product.

There exists a variety of correlations for predicting GCV from ultimate analysis of fuel. The oldest one is Dulong formula (Buckley, 1991) as described below:

$$GCV = 0.336\,(C) + 1.418\,(H) + 0.094\,(S) - 0.145\,(O)$$

In the above equation, the unit of GCV is in mega Joule per kilo gram (mJ/kg, db) while C, H, S, and O are in mass percent of the elements in the sample on a dry basis (db).

The Dulong formula has been fine tuned by Lloyd and Davenport, Tillman, and Boie (Buckley, 1991) into the following formulas:

GCV = 0.3578 (C) + 1.1357 (H) + 0.059 (N) + 0.1119 (S) - 0.0845 (O) by Lloyd and Davenport

GCV = 0.437 (C) - 1.67 by Tillman

GCV = 0.3515 (C) + 1.1617 (H) + 0.06276 (N) + 0.1046 (S) - 0.1109 (O) by Boie

In 1980, Mason and Gandhi used regression analysis and data from 775 USA coals (with less than 30% dry ash) to develop an empirical equation that estimated the calorific value of coal from C, H, S and ash (all on dry basis); their equation was expressed as follows:

GCV = 198.11 (C) + 620.31 (H) + 80.93 (S) + 44.95 (A) – 5153

In the above equation, GCV was in Btu/lb on the dry basis, and C, H, S, O, N, and A are the respective contents of carbon, hydrogen, sulfur, oxygen, nitrogen, and ash in weight percent, also on the dry basis. The formula was claimed to give satisfactory results on higher temperature chars but it needed bias correction for pretreated coal (Mason & Gandhi, 1980). Correlations to calculate calorific value based on elemental analyses has been developed also to estimate heating value of other solid fuels such as biomass (Demirbas, 1997; Friedl et al., 2005; Huang et al., 2008; Sheng and Avecedo, 2005) and refused derived fuel (Buckley and Domalski, 1988).

There exist some GCV equations based on proximate analysis of solid fuels. Majumder et al.(2008) proposed a GCV equation as follows:

GCV = - 0.03(Ash) – 0.11(M) + 0.33(VM) + 0.35(FC)

The GCV unit was in mJ/kg. The analysis basis of ash, moisture (M), volatile matter (VM) and fixed carbon (FC) were air dried basis (adb). They used 250 Indian coal samples and multiple linear regression methods to develop the formula. The average absolute error between the experimental and the predicted data was found to be 1.49% (Majumder et al., 2008). They also reviewed the following correlation developed by Kucukbayrak et al.(1991):

GCV = 76.56 – 1.3(VM + ash) + 0.0073$(VM + ash)^2$

In the above equation, VM and ash were in weigh percentage on dry basis and GCV was determined in mJ/kg also in dry basis. Kucukbayrak equation assumed that GCV has non linear correlation with VM and ash.

A simple equation based on proximate analysis was presented by Cordero et al. (2001) as follows:

GCV = 354.3(FC) + 170.8(VM)

Here GCV was expressed in kJ/kg (db), VM and FC were in weight percent on dry basis. This equation showed the dependence of calorific value on fixed carbon and volatile matter only and it had been derived from multiple linear regression analysis using least square-fitting programme (Cordero et al., 2001). It may be observed from the equation that the ratio of FC to VM coefficient was larger than 2 (354.3/170.8) and it was larger than ratio of FC to VM coefficient derived from Majumder equation (0.35/0.33).

Parikh et. al. (2005) developed general correlation for estimating GCV on dry basis as follows;

GCV = 0.3536(FC) + 0.1559(VM) – 0.0078(ash)

where FC, VM and ash were in weight percent on dry basis and the unit of GCV was on mJ/kg. The correlation termed as ‘general’ since it had been derived based on a large number of data points having widely varying proximate compositions and encompassing all categories of solid carbonaceous materials including coals, lignite, all types of biomass material, and char (Parikh et. al. 2005).

Akkaya (2009) used multiple nonlinear regressions to develop several models for estimation of GCV from the results of proximate analysis of Turkish low rank coals. Among the models, the three models below were proved to give highest degree of correlation.

GCV = 33.078 – 0.72(M) + 0.012(M2) – 1.163(M3) – 0.324(ash2)

GCV = 0.561(M-6.137)(VM0.381)(FC 0.666)

GCV = 0.836(M-8.155)(ash-3.559) (VM 0.35)(FC 0.626)

Although the above selected models predicted GCV rather accurately, Akkaya found that the model involving four independent variables (M, A, VM, FC) provided the most accurate estimation of GCV (R^2 is 0.97). The unit of energy used in the model was mJ/kg and all the variables in the equation were in as received basis (Akkaya, 2009).

Mesroghli et al. (2009) compared linear regression with artificial neural network (ANN) method to develop their correlation. A total of 4540 coal data obtained from U.S. Geological Survey Coal Quality data base were used. In the database, the value of proximate and calorie were in as received basis. They concluded that the accuracy of ANN models was not better or much different than multivariable regression equations.

METHODOLOGY

In this study, all data of proximate analysis and GCV of coal (adb) were taken from data base of coal analysis laboratory of R&D Centre for Coal and Mineral Technology, Ministry of energy and Mineral Resources, Republic of Indonesia. The total number of the data was 451 units. The qualitative correlations between proximate analyses and GCV data were first plotted to examine the linearity of the correlation and then four equations that were developed by Parikh et al. (2005), Cordero et al. (2001), Kucukbayrak et al.(1991) and Majumder et al (2008) were evaluated to understand the suitability of the above equation for estimation the GCV of Indonesian coal. Equation developed in as received basis was not evaluated due to limited number of total moisture data in our data base.

New GCV formulas based on proximate analysis data of Indonesian coal were generated using multiple linear regression method. The basic model of the linear regression was as follows.

$Y = \beta_0 + \beta_1 X_1 + \beta_2 X_2 + \beta_3 X_3 \pm \beta_4 X_4$

Where Y is calculated GCV in calorie/gram, β_0 is constanta, $\beta_{1\text{-}4}$ are regression coefficients and X1-4 are data from proximate analysis i.e. moisture (M), ash, volatile matter (VM) and fixed carbon (FC) in weight percent on air dried basis. The regression coefficient can be calculated manually using matrix method. Below is an example of the calculation.

Suppose the content of VM and FC and measured GCV of coal A and B are as follows:

Coal	VM (%, adb)	FC (%, adb)	GCV (kkal/kg, adb)
A	40	35	4700
B	38	45	5640

If the GCV only depends on two independent variables (VM and FC) only then:

$\beta_1(40) + \beta_2(35) = 4700$
$\beta_1(38) + \beta_2(45) = 5640$

We can write the above data as matrix below:

$$\begin{pmatrix} 40 & 35 \\ 38 & 45 \end{pmatrix}\begin{pmatrix} \beta_1 \\ \beta_2 \end{pmatrix} = \begin{pmatrix} 4700 \\ 5640 \end{pmatrix}$$

where β_1 and β_2 are coefficient for VM and FC respectively

if $\begin{pmatrix} 40 & 35 \\ 38 & 45 \end{pmatrix}$ and A-1= inverse matrix A then

$$A^{-1} * \begin{pmatrix} 40 & 35 \\ 38 & 45 \end{pmatrix}\begin{pmatrix} \beta_1 \\ \beta_2 \end{pmatrix} = A^{-1} * \begin{pmatrix} 4700 \\ 5640 \end{pmatrix}$$

$$\begin{pmatrix} \beta_1 \\ \beta_2 \end{pmatrix} = A^{-1} * \begin{pmatrix} 4700 \\ 5640 \end{pmatrix}$$

$$\begin{pmatrix} \beta_1 \\ \beta_2 \end{pmatrix} = \begin{pmatrix} 0.0957 & -0.0744 \\ -0.0808 & 0.0851 \end{pmatrix} * \begin{pmatrix} 4700 \\ 5640 \end{pmatrix}$$

We get $\beta_1 = 30$ $\beta_2 = 100$ and we found GCV equation as follows

GCV= 30 (VM) + 100 (FC)

The matrix calculation is getting complicated if many coefficients must be calculated. In this study the regression coefficient and constanta were determined through matrix computation using SPSS software. SPSS is among the most widely used programs for statistical analysis.

Equations obtained in this analysis were compared on the basis of coefficient of multiple determinations or R-squared (R^2). R-squared is a statistical measure of how close the data are to the fitted regression line. The higher R^2 value of the equation the better is the estimation (a perfect correlation has a R^2 of 1).

Ten (10) equations that will be developed have pattern listed in Table 1. It includes three (3) equations with one independent variable, four (4) equations with two independent variables, two (2) equations with three independent variables and one (1) equation having four independent variables. The 10 equations will be rank based on their R^2 or R-squared value.

RESULTS AND DISCUSSIONS

Correlation between Measured GCV and the Result of Proximate Analyses

Proximate analysis measures the percentage by weight of moisture fixed carbon, volatiles matter, and ash in coal. Moisture is measured from the weight loss of the coal when heated to 110°C for 1 hour. Ash is measured from the weight percentage of material remaining after the coal sample is completely incinerated. Volatile matter is the weight percent loss when the coal sample is heated in a reducing atmosphere (out of contact with air) to 900°C and fixed carbon is calculated by subtracting the sum of moisture, ash and volatile matter from 100 percent.

Figure 1 presents qualitative correlation between GCV and proximate analysis data. It was observed that ash and M have negative effects on GCV, on the other hand, FC and VM contents both have positive effects on the GCV. Thus, the amounts of FC and VM directly contribute to the heating value of coal. Strongest correlation with the value of R^2 of 0.803 was found between GCV and fixed carbon content of coal (Figure 1d). The trend line of the data and its equation are also presented in Figure 1. Since the gradient of the trend line in Figure 1d is larger than that of Figure 1c, it can be concluded that fixed carbon acts as a main heat generator during burning.

It is predicted that a GCV correlation may give higher R Squared value if GCV is plotted to added value of M and ash content or to added value of both VM and FC since both ash and moisture have negative effect on GCV and both FC and VM have positive effect on GCV. Figure 2a shows correlation between GCV and added value of both moisture and ash. Correlation between GCV and added value of moisture and ash give much higher R-squared value than correlation between GCV and moisture only (Figure 1a) or between GCV and ash only (Figure 1b) as predicted. Coal ash is formed by the combustion of mineral matter in coal. The mineral matters often found in coals are aluminum silicate, quartz, pyrite and calcium carbonate. Pyrite released heat and carbonate absorbed heat during combustion but the main component of ash (alumina silicate) did not react with oxygen to produce or absorb heat during coal combustion at low temperature. It may be assumed that mineral matter in coal did not release heat during combustion and as result the GCV of coal decreased when ash content increased. Opposite result appears when GCV is correlated with the sum of fixed carbon and volatile matter (figure 2b). It gives R-squared value lower than that of correlation between GCV and fixed carbon only

Table 1. List of GCV equations to be developed and their independent variables

No	Independent variable	Equation
1	VM	GCV = β0 + β1(VM)
2	FC	GCV = β0 + β1(FC)
3	(VM+FC)	GCV = β0 + β1(VM + FC)
4	VM & FC	GCV = β0 + β1(VM) + β2(FC)
5	M & Ash	GCV = β0 + β1(M) + β2(Ash)
6	(M+Ash) & FC	GCV = β0 + β1(M + Ash) + β2(FC)
7	(M+Ash) & (VM+FC)	GCV = β0 + β1(M + Ash) + β2(VM + FC)
8	M, VM & FC	GCV = β0 + β1(M) + β2(VM) + β3(FC)
9	Ash, VM & FC	GCV = β0 + β1(Ash) + β2(VM) + β3(FC)
10	M, Ash, VM & FC	GCV = β0 + β1(M) + β2(Ash) + β3(VM) + β4(FC)

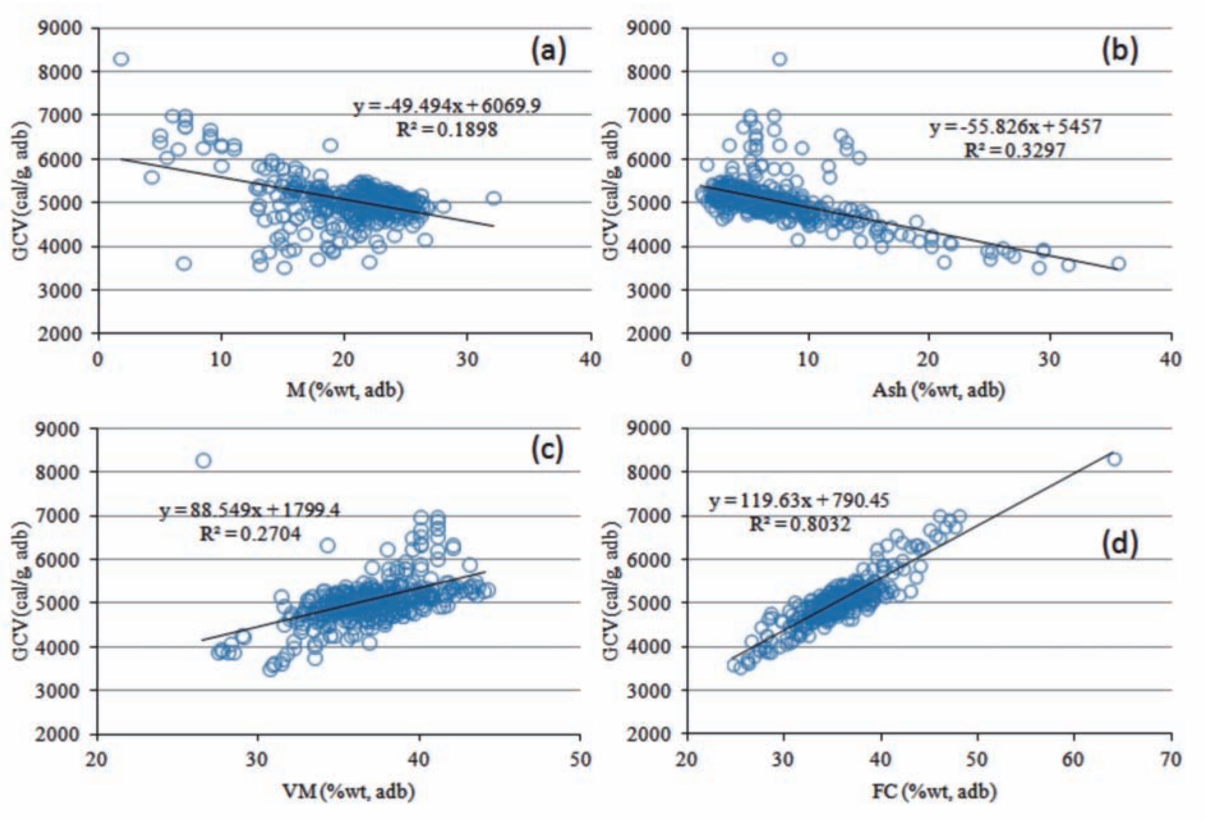

Figure 1. Correlation between GCV and individual proximate analyses data

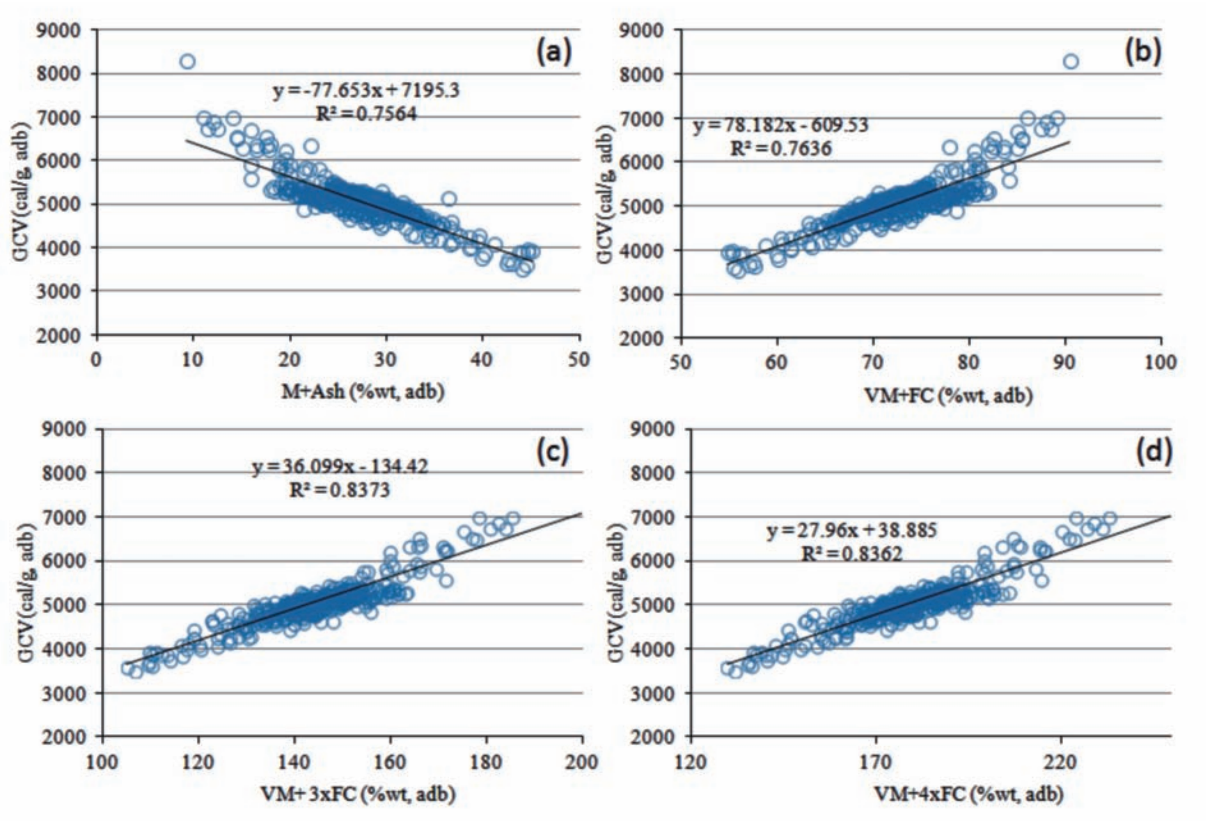

Figure 2. Correlation between GCV and compounded data from proximate analyses

(Figure 1d). Volatile matter contains many kind of gas and liquids and the heating value of VM also depends on coal rank (Urkan and Arikol, 1989). Thus the contribution of VM to GCV not only depends on percentage of VM but also the calorific value of VM. Therefore, an attempt to correlate GCV with percentage of VM only will not give high R-squared value.

Based on equation developed by Cordero et al. (2001) and Majumder et al. (2008) the contribution of VM and FC to GCV was not equal. Since fixed carbon was the main heat generator during burning, the coefficient of FC must be larger than that of volatile matter. Figure 2c and Figure 2d present the correlation of GCV if coefficient of FC is 3 and 4 times larger than VM, respectively. The R-squared value reached optimum when the coefficient of fixed carbon is in between 3 and 4 times larger than that of volatile matter.

Further, more accurate correlation of GCV may be obtained if the heating value of VM is considered. Urkan and Arikol (1989) had measured the heating values of 20 kinds of Turkish coal volatile matter. The heating value of VM vary from 17,000 to 26,000 kJ/kg for lignites and from 30,000 to 38,000 kJ/kg for subbituminous and bituminous coals. They did not report VM with heating value in the range of 26,000 to 30,000 kJ/kg. A larger number of coal samples should be analyzed to obtain comprehensive data of volatile matter heating value.

Goutal (1902) quoted by Majumder (2008) developed a correlation that consider the rank of coals as follows :

$$GCV = 82\ FC + a\ VM$$

Here FC and VM denotes fixed carbon and volatile matter respectively and 'a' is a constant that depends on coal rank. This correlation may accurately predict the GCV but it will limit the use of the equation to a certain coal rank only. Therefore in this paper the calorific value of VM is not considered in developing the GCV equations.

The Use of Some Published Equations for Calculating GCV of Indonesian Coal

Figure 3a, 3b, 3c and 3d present correlation between measured GCV and calculated GCV using formulas developed by four group of researchers (Parikh et al., 2005; Cordero et al., 2001; Kucukbayrak et al., 1991; Majumder et al, 2008). Among the four equations, the equation developed by Kucukbayrak showed the lowest R-squared value. Kucukbayrak equation assumed that GCV has non linear correlation with

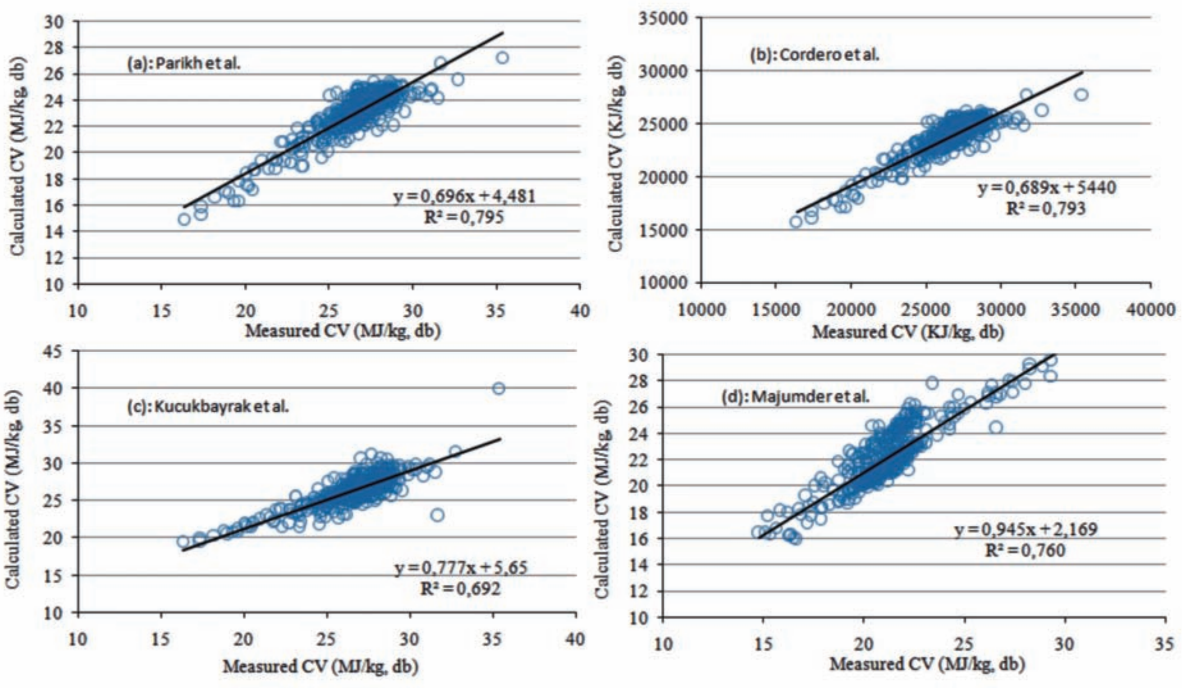

Figure 3. Aplicabilty of existing equation for calculating GCV of Indonesian coal

VM and ash. Based on Mesroghli work, the non linear equation of GCV are not better than the multivariable regression equations (Mesroghli et al., 2009).

The R-squared value of Majumder is lower than that of Parikh and Cordero. It is may be explained by the coefficient of FC and VM on the equations. The coefficient ratio of FC/VM is more than double for Parikh and Cordero equations while it was about 1 for Majumder equation. The GCV equation for Indonesian coal will result in high R-squared value if the coefficient of fixed carbon is much larger than the coefficient of volatile matter (Figure 2c and 2d).

Development of New GCV Equation for Indonesian Coals

Table 2 presents new equations developed using proximate data and GCV value of Indonesian coals. Equation No. 10 which use VM only as independent variable showed the lowest R-Squared value while the R-Squared value was moderate when FC was used as independent variable. Based on their R-squared value, equation No. 1-6 gave better correlation than equation developed by four group of researchers (Parikh et al., 2005; Cordero et al., 2001; Kucukbayrak et al., 1991; Majumder et al, 2008). It seems that the four equations may be best suited only for coals that are used for the study. The best correlation (equation No. 1) involving four independent variables. Based on Akkaya (2009) equation involving four independent variables (M, A, VM, FC) provides the most accurate estimation of GCV.

Measured GCV and calculated GCV using Equation No. 1 were plotted in Figure 4. It is remarkable to note that the use of the new equation (present work) result in calculated GCV with smaller difference than measured GCV. Thus, it is suggested to use the new equation developed here for estimating the GCV of Indonesian coal. In addition, the new developed GCV equations uses GCV unit of coals (cal/g) that more commonly used in Indonesia.

CONCLUSIONS

Equations for estimating GCV of Indonesian coals based on proximate analysis data coal have been developed and the existing GCV equations in the literature have been reviewed. The following conclusions were obtained from the present work:

a. GCV of Indonesian coal has stronger correlation with FC than that of with VM, therefore, the coefficient of FC should be larger that that of VM in the GCV equations.

b. The best GCV equation for Indonesian coal has the following form: GCV= 25.284 (M) + 30.572 (ash) + 62.127 (VM) + 138.117 (FC) - 2890.095. GCV unit is in calorie/gram (adb) and M, ash, VM and FC are in weight percent (adb).

c. The new developed equation has been used to calculate the GCV of Indonesian coal and it gave more accurate result than that of existing GCV equation in the literatures. Therefore, it is suggested to use the new equation for estimating the GCV of Indonesian coal.

Table 2. List of new developed GCV equations and their R-squared value

No.	Formula	R Squared
1	GCV= 25.284 (M) + 30.572 (Ash) + 62.127(VM) + 138.117 (FC) - 2890.095	0.839
2	GCV= -4.718 (M) + 31.960 (VM)+ 107.694 (FC) + 135.661	0.838
3	GCV= 5.167 (Ash) + 36.891 (VM) + 112.726 (FC) - 358.420	0.838
4	GCV= 34.208 (VM) + 109.172(FC) - 96.051	0.837
5	GCV= -32.755 (M + Ash) + 76.875 (FC) + 3217.083	0.835
6	GCV= 119.634 (FC) + 790.445	0.803
7	GCV= 17.175 (M + Ash) + 95.335 (VM + FC) - 2325,677	0.764
8	GCV= 78,182 (VM+FC)-609,534	0.763
9	GCV= -78.314 (M) + -77.235(Ash) - 7205.764	0.756
10	GCV= 88.549 (VM) + 1799.352	0.270

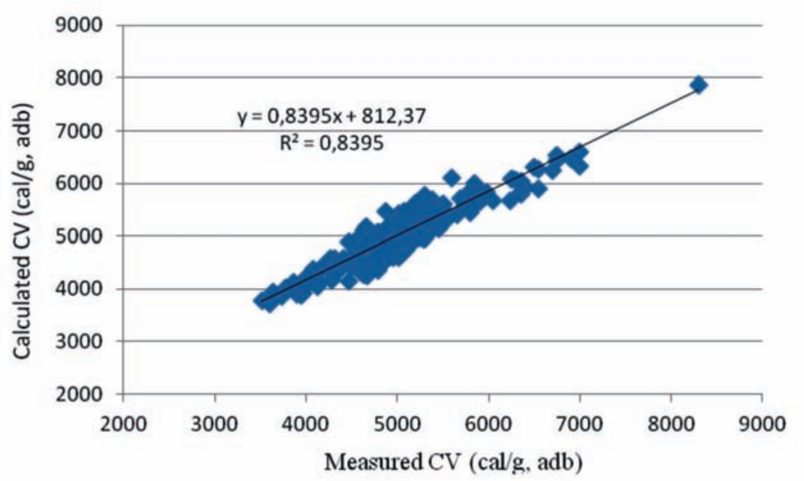

Figure 4. Correlation between measured GCV and calculated GCV using equation No.1

ACKNOWLEDGEMENTS

The author wish to thank to Mrs. Tuti Hernawati for teaching the SPSS software and to head of R&D Centre for Coal and Mineral Technology, Ministry of energy and Mineral Resources, Republic of Indonesia for allowing to use the data.

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