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THE GROSS CALORIFIC VALUE IN THE WET ASH-FREE STATE

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Abstract

Analysis of 63 samples of coal concentrates (from Ukraine, Russia, the United State, Canada, Australia, Poland) currently employed at Ukrainian coke plants permits the development of formulas predicting the gross calorific value in the wet ash-free state. Three separate formulas are derived (for the Ukrainian coal, the Russian coal, and the other imported coal), with allowance for the differences in the petrographic composition and degree of reduction of the coal. The prediction is based on the volatile matter, the mean vitrinite reflection coefficient, and the sum of fusinized components. The accuracy of the predictions is within the standard tolerances (σ≤0.3 MJ/kg).

Keywords: coal; gross calorific value; mathematical equations; statistical estimates.

1. Introduction

In previous research, 31 samples of coal concentrate produced in Ukraine, Russia, the United States and Poland used at Ukrainian coke plants were studied [1–3].That research culminated in the formulation of mathematical models permitting the prediction of the gross calorific value Q_s^{af}, in the wet ash-free state on the basis of the coal properties.

Those formulas permitted the prediction of Q_s^{af} with high accuracy. The mean square deviation of the actual values from the calculation results was within the permissible standard error in determining Q_s^{af} (σ≤0.3 MJ/kg). On the basis of the formulas, a method for determining Q_s^{af} was developed and is still used by Ukrainian coke plants to evaluate imported coal [4].

However, Ukrainian coke plants now employ a broader range of coal concentrates than were considered in [1–3]. Accordingly, in the present work, we analyze the relation between the properties of the coal now employed and its gross calorific value Q_s^{af}, in the wet ash-free state.

2. Experimental

We consider 63 coal samples: 21 from Ukraine; 21 from Russia; and 21 imported from elsewhere (the United State, Canada, Australia and Poland). Since the oxidation of the coal on its quality is significantly oxidized coal is excluded from the samples [5-6].

Table 1 present the maximum, minimum and mean value of the coal properties for each group and also for the complete set of coal samples. The mean of volatile matter is approximately the same for all three groups (V^{daf} =28.7–32.1 %). That indicates similar variation in all the groups.

Coal of both low sulfur content (S_t^d = 0.25 %) and high sulfur content (S_t^d = 3.92 %) is presented. The maximum moisture content W_{max} fluctuates widely: from 1.3 % to 5.9 %. The gross calorific value Q_s^{daf} in the dry ash-free state varies from 33.42 to 36.58 MJ/kg; the gross calorific value Q_s^{af} in the wet ash-free state varies from 31.40 to 35.92 MJ/kg.

Table 2 present the maximum, minimum and mean value of the coal samples petrographic properties.

Table 1. Technological properties of the three groups of coal

Group	Value	Proximate analysis, %				Maximum moisture content, %	Gross calorific value, MJ/kg	
		W^a	A^d	S_t^d	V^{daf}	W_{max}	Q_s^{daf}	Q_s^{af}
Ukrainian coal	Max	2.2	9.8	3.92	42.9	4.5	36.36	35.51
	Min	0.5	3.7	0.73	26.2	1.7	33.82	32.24
	Mean	1.1	7.2	1.94	32.1	2.5	35.67	34.72
Russian coal	Max	2.7	10.5	2.49	41.3	5.9	36.50	35.89
	Min	0.4	4.8	0.25	19.8	1.5	33.42	31.40
	Mean	1.4	7.9	0.67	30.9	3.3	35.37	34.13
Other imported coal *(United States, Canada, Australia, and Poland)*	Max	1.2	9.8	1.25	34.5	2.3	36.58	35.92
	Min	0.4	5.3	0.39	18.7	1.3	35.59	34.72
	Mean	0.8	8.4	0.79	28.7	2.0	35.99	35.20
All the coal	Max	2.7	10.5	3.92	42.9	5.9	36.58	35.92
	Min	0.4	3.7	0.25	18.7	1.3	33.42	31.40
	Mean	1.1	7.8	1.14	30.6	2.6	35.68	34.68

Table 2. Petrographic characteristics of the three groups of coal

Group	Value	Mean vitrinite reflection coefficient, %	Petrographic composition, %				
		R_o	*Vt*	*Sv*	*I*	*L*	*ΣFC*
Ukrainian coal	Max	1.20	92	1	32	10	32
	Min	0.65	65	0	5	0	6
	Mean	1.02	83	0	14	3	15
Russian coal	Max	1.56	71	1	27	1	26
	Min	0.66	71	1	27	1	26
	Mean	1.00	71	1	27	1	26
Other imported coal	Max	1.50	88	2	42	6	44
	Min	0.90	53	0	8	0	9
	Mean	1.13	75	1	22	2	23
All the coal	Max	1.56	95	4	64	10	66
	Min	0.65	33	0	4	0	4
	Mean	1.05	76	1	21	2	21

The metamorphic development of the coal, characterized by the vitrinite reflection coefficient R_o, varies from 0.65 to 1.56 %. The petrographic uniformity is petrographic greatest for Ukrainian coal: Vt_{me} = 83 %, as against 71 % for Russian coal. The mean sum of fusinized components is 15 % for Ukrainian coal, 26 % for Russian coal, and 23 % for the other imported coal.

Table 3 present the maximum, minimum, and mean value of the coal samples ultimate composition (C^{daf}, H^{daf}, N^{daf}, $S_t^{d,}$ and O_d^{daf}) and the structural parameters (C_{ar}, δ, f_a, cA). The structural parameters are determined from the formulas in [7]. The mean content of carbon, hydrogen and oxygen is relatively similar for the three groups of coal (Table 3). Ukraine coal is characterized by the largest total sulfur content ($S^d{}_{t\,me}$ = 3.92 %) and the smallest nitrogen content ($N^{daf}{}_{me}$ = 1.76 %), and Russian coal by the smallest total sulfur content ($S^d{}_{tme}$ = 0.67 %) and the largest nitrogen content ($N^{daf}{}_{me}$ =2.27 %). For the other imported coal $S^d{}_{t\,me}$=0.79 % and $N^{daf}{}_{me}$=1.59 %. Turning to the structural parameters, we note that the content of aromatic carbon C_{ar} is least for the Ukrainian coal (21.47 %), higher for the Russian coal (23.34 %), and highest for the other imported coal (25.44 %). The same pattern is observed for the aromatic structure f_a of the organic mass: 0.693, 0.696, and 0.706, respectively. The degree of molecular association cA and nonsaturation δ of unit organic mass also increase in the same sequence: 0.769, 0.775, and 0.785 for cA; and 8.918, 9.106 and 9.310 for δ. The foregoing indicates that the molecular structure of the coal is somewhat different for the three groups: the condensation of the organic mass is greatest for the non-Russian important coal.

Table 3. Ultimate composition and structural parameters of the three groups of coal

Group	Value	Ultimate composition, %					Structural parameters			
		C^{daf}	H^{daf}	N^{daf}	S^d_t	O_d^{daf}	C_{ar}, %	δ	f_a	cA
Ukrainian coal	Max	88.62	6.22	2.15	3.92	9.30	26.48	9.596	0.724	0.796
	Min	78.93	5.21	1.31	0.73	2.44	12.74	7.052	0.609	0.693
	Mean	86.11	5.56	1.76	1.94	4.62	21.47	8.918	0.693	0.769
Russian coal	Max	91.14	6.17	3.16	2.49	9.11	34.97	10.158	0.741	0.816
	Min	82.41	4.99	1.09	0.25	2.17	15.93	7.920	0.640	0.730
	Mean	86.71	5.51	2.27	0.67	4.83	23.34	9.106	0.696	0.775
Other imported coal	Max	90.76	6.27	2.35	1.25	6.47	33.40	10.235	0.47	0.817
	Min	85.9	4.92	1.22	0.39	2.13	20.71	8.359	0.658	0.751
	Mean	88.00	5.47	1.59	0.79	4.19	25.44	9.310	0.706	0.785
All the coal	Max	91.14	6.27	3.16	3.92	9.30	34.97	10.235	0.747	0.817
	Min	78.93	4.92	1.09	0.25	2.13	12.74	7.052	0.609	0.693
	Mean	86.94	5.51	1.87	1.14	4.55	23.42	9.111	0.698	0.776

3. Results and discussion

In calculating the pair correlation coefficients between the properties of the coal and the gross calorific value Q_s^{af} in the wet ash-free state (Table 4), we take account of the relation between the coal properties and their calorific value from [8-9].

Table 4. Pair correlation coefficients between the properties of the coal and the gross calorific value and corresponding $|r|\sqrt{n-1}$ values

Property	W^a	A^d	S^d_t	V^{daf}	W_{max}	R_o	Vt
r	-0.950	0.119	0.244	-0.675	-0.937	0.756	-0.027
$\lvert r\rvert\sqrt{n-1}$	7.480	0.937	1.921	5.315	7.378	5.953	0.213
Property	Sv	I	L	ΣFC	C^{daf}	H^{daf}	N^{daf}
r	0.057	0.065	-0.290	0.067	0.790	-0.521	-0.464
$\lvert r\rvert\sqrt{n-1}$	0.449	0.512	2.283	0.528	6.220	4.102	3.654
Property	O_d^{daf}	Q_s^{daf}	cA	fa	Car	δ	
r	-0.833	0.978	0.730	0.704	0.709	0.692	
$\lvert r\rvert\sqrt{n-1}$	6.559	7.701	5.748	5.543	5.583	5.449	

The significance of correlation coefficients is verified by comparing the absolute value of the product of $|r|\sqrt{n-1}$ with its critical value (H) at the specified reliability level (P) [10]. With P=0.999, the critical value H is 3.183 for the 63 samples.

Table 4 presents the values of the product $|r|\sqrt{n-1}$ for all the correlation of interest. Comparing the actual and critical values indicate that with 0,999 probability, Q_s^{af} is correlated with W^a, V^{daf}, W_{max}, R_0, C^{daf}, O_d^{daf}, Q_s^{daf}, cA, f_a, C_{ar} and δ.

Analysis of table 5 indicates that satisfactory prediction of Q_s^{af} (with permissible discrepancy ($\sigma \leq 0.3$ MJ/kg) is only possible of the basis of W^a, W_{max}, Q_s^{daf} for witch (δ=*0.29; 0.26* and *0.20 MJ/kg* respectively). For the other properties (R_0, C^{daf}, O_d^{daf}, cA, f_a, C_{ar}, and δ), σ=0.39–0.41 MJ/kg.

When using V^{daf} for prediction, we find that (σ=0.34 MJ/kg), which is lower but still exceeds the acceptable upper limit of *0.3 MJ/kg*.

On W_{max} and Q_s^{af} makes sense, since these characteristics are used to calculate Q_s^{af} in accordance with ISO 1170:77 [11]. Likewise, we know that W^a is closely related to W_{max}, which provides good predictions of Q_s^{af}. The relationship may be assessed in terms of the determination coefficients. Since the formulas and statistical characteristics are obtained for all the coal samples, the mean square deviation of the calculated results from the actual data may usefully be calculated for each of the groups (Table 5).

Table 5. Mathematical equations and statistical characteristics for the relation between Q_s^{af} and the properties of the three groups of coal

Eq.	Formula	Statistical characteristics		σ, MJ/kg			
		r	D,%	All the coal	Ukrainian coal	Russian coal	other imported coal
1	$Q_s^{af} = 1.7501 \cdot W^a + 36.65$	0.95	90.2	0.29	0.30	0.36	0.23
2	$Q_s^{af} = -0.938 \cdot W_{max} + 37.10$	0.96	91.3	0.26	0.27	0.31	0.19
3	$Q_s^{af} = -0.007 \cdot (V^{daf})^2 + 0.363 \cdot V^{daf} + 31.05$	0.85	71.4	0.34	0.37	0.51	0.34
4	$Q_s^{af} = -4.823 \cdot (Ro)^2 + 13.32 \cdot Ro + 26.28$	0.84	70.6	0.43	0.36	0.56	0.35
5	$Q_s^{af} = -0.0503 \cdot (C^{daf})^2 + 9{,}0512 \cdot C^{daf} - 371.45$	0.89	79.4	0.41	0.36	0.57	0.32
6	$Q_s^{af} = -0.076 \cdot ({O_d}^{daf})^2 + 0.366 \cdot {O_d}^{daf} + 34.84$	0.90	80.5	040	0.28	0.50	0.41
7	$Q_s^{af} = 1.459 \cdot {Q_s}^{daf} - 17.36$	0.98	95.6	0.20	0.15	0.29	0.10
8	$Q_s^{af} = -502.4 \cdot (cA)^2 + 805.4 \cdot cA - 287.4$	0.80	64.6	0.39	0.34	0.55	0.31
9	$Q_s^{af} = -189.4 \cdot (fa)^2 + 279.7 \cdot fa - 68.00$	0.81	65.6	0.39	0.31	0.52	0.36
10	$Q_s^{af} = -0.013 \cdot (Car)^2 + 0.774 \cdot \mathrm{Car} + 24.12$	0.88	77.2	0.39	0.30	0.59	0.35
11	$Q_s^{af} = -0.363 \cdot (\delta)^2 + 7.317 \cdot \delta - 1.49$	0.81	64.9	0.40	0.34	0.58	0.30

Analysis shows that the prediction of Q_s^{af} on the basis of the proposed equations is worst for the Russian coal: the mean square deviation of the calculation results from the actual data exceeds the permissible error (0.3 MJ/kg). For the Ukrainian coal and other imported coal, the error is smaller; it is approximately the same for both groups. This may be attributed to petrographic inhomogeneity of the Russian coal [12].

On the basis of the foregoing, it is expedient to develop mathematical equations for each of the groups: Ukrainian coal, Russian coal and other imported coal. The Russian coal is considered separately on account of its petrographic inhomogeneity. The Ukrainian coal and the other imported coal are considered separately on the basis that Donetsk and United State coal of the same metamorphic development are known to differ significantly in composition (C^{daf}, H^{daf}, O^{daf}), structural characteristics (C_{ar}, f_a, δ and B) and technological properties (R_0, V^{daf}, y, Q_s^{daf}). The differences are mainly due to the different degrees of reduction of the coal samples [7].

Tables 6–8 present equations and statistical characteristics analogous to those in Table 5, but for the individual groups of coals. Analysis of Tables 6–7 indicates that the formulas for the individual groups permit significantly more reliable predictions. In particular, the mean square deviation of the calculation results from the actual data is lower.

Table 6. Mathematical equations and statistical characteristics for the relation between Q_s^{af} and the properties of Ukrainian coal

Eq.	Formula	Statistical characteristics		
		r	D, %	σ, MJ/kg
12	$Q_s^{af} = -1.731 \cdot W^a + 36.64$	0.91	83.7	0.29
13	$Q_s^{af} = -1.002 \cdot W_{max} + 37.19$	0.94	87.6	0.26
14	$Q_s^{af} = -0.015 \cdot (V^{daf})^2 + 0.891 \cdot V^{daf} + 22.05$	0.90	81.2	0.22
15	$Q_s^{af} = -12.58 \cdot (Ro)^2 + 28.15 \cdot Ro + 19.31$	0.91	83.1	0.30
16	$Q_s^{af} = -0.061 \cdot (C^{daf})^2 + 10.861 \cdot C^{daf} - 448.22$	0.82	67.8	0.27
17	$Q_s^{af} = -0.049 \cdot ({O_d}^{daf})^2 + 0.128 \cdot {O_d}^{daf} + 35.30$	0.95	90.5	0.23
18	$Q_s^{af} = 1.370 \cdot {Q_s}^{daf} - 14.14$	0.98	96.3	0.14
19	$Q_s^{af} = -346.6 \cdot (cA)^2 + 558.6 \cdot cA - 189.7$	0.72	51.6	0.34
20	$Q_s^{af} = -151 \cdot (fa)^2 + 227.9 \cdot fa - 50.62$	0.90	81.9	031
21	$Q_s^{af} = -0.0128 \cdot (Car)^2 + 0.7458 \cdot \mathrm{Car} + 24.749$	0.93	87.1	0.27
22	$Q_s^{af} = -0.2942 \cdot (\delta)^2 + 6.0919 \cdot \delta + 3.8819$	0.89	80.1	0.33

Table 7. Mathematical equations and statistical characteristics for the relation between Q_s^{af} and the properties of Russian coal

Eq.	Formula	Statistical characteristics		
		r	*D,%*	*σ, MJ/kg*
23	$Q_s^{af} = -1.766 \cdot W^a + 36.64$	0.96	91.3	0.36
24	$Q_s^{af} = -0.976 \cdot W_{max} + 37.33$	0.97	93.6	0.29
25	$Q_s^{af} = -0.004 \cdot (V^{daf})^2 + 0.159 \cdot V^{daf} + 33.69$	0.84	70.8	0.44
26	$Q_s^{af} = -2.659 \cdot (Ro)^2 + 8.620 \cdot Ro + 28.50$	0.85	72.2	0.52
27	$Q_s^{af} = -0.0378 \cdot (C^{daf})^2 + 6.8722 \cdot C^{daf} - 276.96$	0.91	83.7	0.40
28	$Q_s^{af} = -0.058 \cdot ({O_d}^{daf})^2 + 0.144 \cdot {O_d}^{daf} + 35.10$	0.93	87.4	0.41
29	$Q_s^{af} = 1.428 \cdot {Q_s}^{daf} - 16.4$	0.98	95.4	0.26
30	$Q_s^{af} = -476.7 \cdot (cA)^2 + 766.0 \cdot cA - 272.5$	0.85	72.4	050
31	$Q_s^{af} = -126.5 \cdot (fa)^2 + 191.9 \cdot fa - 37.65$	0.77	59.4	0.48
32	$Q_s^{af} = -0.0094 \cdot (Car)^2 + 0.6123 \cdot \mathrm{Car} + 25.518$	0.89	78.4	0.50
33	$Q_s^{af} = -0.2746 \cdot (\delta)^2 + 5.724 \cdot \delta + 5.3076$	0.77	59.3	0.48

Table 8. Mathematical equations and statistical characteristics for the relation between Q_s^{af} and the properties of the other imported coal

Eq.	Formula	Statistical characteristics		
		r	D, %	σ, MJ/kg
34	$Q_s^{af} = 1.291 \cdot W^a + 36.28$	0.80	63.4	0.20
35	$Q_s^{af} = -1.093 \cdot W_{max} + 37.37$	0.85	72.0	0.18
36	$Q_s^{af} = -0.0003 \cdot (V^{daf})^2 - 0.0305 \cdot V^{daf} + 36.29$	0.71	50.7	0.24
37	$Q_s^{af} = 0.268 \cdot (Ro)^2 + 0.669 \cdot Ro + 34.09$	0.73	52.7	0.23
38	$Q_s^{af} = -0.027 \cdot (C^{daf})^2 + 4.9292 \cdot C^{daf} - 189.32$	0.72	51.9	0.25
39	$Q_s^{af} = -0.021 \cdot ({O_d}^{daf})^2 + 0.024 \cdot {O_d}^{daf} + 35.71$	0.75	56.4	0.22
40	$Q_s^{af} = 1.283 \cdot {Q_s}^{daf} - 11$	0.98	95.3	0.10
41	$Q_s^{af} = -57.92 \cdot (cA)^2 + 103.3 \cdot cA - 10.24$	0.61	37.4	0.28
42	$Q_s^{af} = -22.16 \cdot (fa)^2 + 40.45 \cdot fa + 17.68$	0.58	33.2	0.28
43	$Q_s^{af} = -0.052 \cdot (Car)^2 + 0.3382 \cdot \mathrm{Car} + 30.06$	0.71	51.1	0.24
44	$Q_s^{af} = -0.0959 \cdot (\delta)^2 + 2.2156 \cdot \delta + 22.9$	0.59	35.4	0.28

The following predictions of Q_s^{af} are acceptable, on the basis that the error should be no more than 0.3 MJ/kg [13]:

–from Eqs. (12) – (18) and (21) for Ukrainian coal;
–from Eqs. (24) and (29) for Russian coal;
–from Eqs. (34) – (44) for of the other imported coal.

Since Q_s^{af} and W_{max} are not routinely monitored at Ukrainian coke plants, they cannot be used for the prediction of Q_s^{af} for Russian coal.

Accordingly, for Russian coal, a formula for predicting Q_s^{af} has been developed in the form of a second-order polynomial in terms of the vitrinite reflection coefficients R_0 and the sum of fusinized components ΣFC, which are routinely determined in coke-plant laboratories

$$Q_s^{af} = 0{,}0321 \cdot R_0 \cdot \Sigma FC - 6{,}134 \cdot R_0^2 + 16{,}5891 \cdot R_0 + 25{,}6885 - 0{,}1304 \cdot \sum FC + 0{,}0011 \cdot \sum FC^2$$

The corresponding determination coefficient *D* is 89.8 %; the multiple-correlation coefficients *r*=0.948; the mean square deviation is 0.29 MJ/kg. Thus, for Russian coal, this formula may be recommended for the prediction of Q_s^{af}. As already noted, R_0 and V^{daf} may be used to predict Q_s^{af} for Ukrainian coal and the other imported coal. The accuracy of the prediction is satisfactory: σ=0.27–0.30 MJ/kg.

Note that the mathematical formulas obtained permit accurate prediction of gross calorific value Q_s^{af} in the wet ash-free state for Ukrainian, Russian and other imported coal supplied to Ukrainian coke plants, in contrast to those in [1–4].

4. Conclusions

1. We have analyzed 63 sample of coal concentrates (from Ukrainian, Russian, the United States, Canada, Australia, and Poland) currently employed at Ukrainian coke plants.
2. We have shown that the gross calorific value Q_s^{af} in the wet ash-free state cannot be accurate prediction from parameters routinely determined in coke-plant laboratories on the basis of a single universal equation, on account of the petrographic inhomogeneity and different degrees of reduction of individual bath components.
3. We have developed predictive equations on the basis of the volatile matter, the mean vitrinite reflection coefficients and the sum of lean microcomponents for Ukrainian coal, for Russian coal and for other imported coal (from the United States, Canada, Australia and Poland). The accuracy of the predictions is within the standard tolerances ($\sigma \leq 0.3$ MJ/kg).

Symbols

Q_s^{af}	*gross calorific value in the wet ash free state, MJ/kg;*
W_{max}	*moisture holding capacity, %;*
Q_s^{daf}	*gross calorific value in the dry ash free state, MJ/kg;*
R_0	*mean vitrinite reflection coefficient, %;*
A^d	*ash content of coal, %;*
V^{daf}	*volatile matter, %;*
W^a	*moisture in the analysis sample, %;*
S_t^d	*content of sulphur, %;*
C^{daf}, H^{daf}, N^{daf}, O^{daf}	*content of carbon, hydrogen, nitrogen and oxygen in the dry ash free state, %;*
C_{ar},	*content of aromatic carbon, %;*
f_a, δ	*structural characteristics;*
ΣLC	*content of fusinized components, %;*
Vt	*content of vitrinite, %;*
I	*content of inertinite, %;*
L	*content of liptinite, %;*
D	*coefficient of determination, %;*
r	*coefficient of correlation;*
σ	*mean square deviation, MJ/kg.*

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