Journal of Radioanalytical and Nuclear Chemistry, Vol. 262, No. 2 (2004) 493–496

Detection of low caloric power of coal by pulse fast-thermal neutron analysis

Gu De-shan,[1] Jing Shi-wei,[1,2]* Sang Hai-feng,[1] Qiao Shuang,[1] Liu Yu-ren,[1] Liu Lin-mao[1]

[1] *Physics Department, Northeast Normal University, Changchun 130024, P.R. China*
[2] *Changchun Institute of Optics, Fine Mechanics and Physics, Chinese Academy of Sciences, Changchun 130033, P.R. China*

(Received March 11, 2004)

This paper introduces the analysis method and principle of pulse fast-thermal neutron analysis (PFTNA). A system for the measurement of low caloric power of coal by PFTNA is also presented. The 14 MeV pulse neutron generator and BGO detector and 4096 MCA were applied in this system. A multiple linear regression method applied to the data solved the interferential problem of multiple elements. The error of low caloric power between chemical analysis and experiment was less than 0.4 MJ/kg.

Introduction

It is often necessary to have accurate data on low caloric power of coal in coal cleaning and power plants. Until recent years, this information was obtained by means of sampling techniques associated with conventional chemical analysis. The chemical methods can only be implemented on small samples. The analysis process is very complex, and needs sampling, weighting, drying at constant temperature, testing and so on. The chemical method cannot direct the industrial product on time because of the long analysis time for a coal sample. Pulsed fast-thermal neutron analysis (PFTNA) is a bulk material analysis technique, which makes it possible to detect elements in coal in a short time. Many institutes are engaged in the development of PFTNA on coal analysis.[1–4]

A fast coal inspection system was developed in the Radiation Technology Institute of Northeast Normal University. The pulsed neutron generator and BGO detector employed in the system were provided by the Northeast Normal University and the Shanghai Silicate Institute, respectively. The MCA with 4096 channels was offered by the Shanghai Atomic Nuclear Institute. A multiple linear regression method applied to deal with data solved the interferential problem of multiple elements. The error of low caloric power ($Q_{\text{net ar}}$) between chemical analysis and experiment was below 0.4 MJ/kg.

Experimental

Principles and method

The reaction between neutrons and a nucleus includes mainly inelastic scattering (n, n', γ) and neutron capture (n, γ). When the 14 MeV neutrons react with the elements in coal the (n, n', γ) reaction takes place over a very short time (10^{-8} to 10^{-7} s) with emitting of characteristic gamma-rays at one time. After several times collision neutrons decrease their energy to become thermal neutrons over 10^{-6} to 10^{-5} seconds. Some thermal neutrons diffuse in the coal by (n, n) reaction and the others are captured by some nucleus to emit characteristic gamma-rays via (n, γ) reactions. It is not easy to detect the pure inelastic spectra according to the reaction time and the actual condition of pulsed neutron generator. A gate circuit is employed in the experiment to separate spectrum I (the overall spectrum of inelastic, capture and activation) and spectrum II (the sum spectrum of capture and activation). The relatively pure inelastic spectrum is obtained by detracting spectrum I to II and it is useful to detect C, O and H in coal.

In quantitative analysis the characteristic peak area of elements is represented by net counts N (accumulated in the measurement period). The relationship between net counts, N and the element content measured, G, is as follows:

$$N = \frac{GN_A}{A}\phi\sigma\varepsilon j\alpha t \qquad (1)$$

where N are net counts of a characteristic peak (in measurement period t), G is the element content, N_A is the Loschmidt constant, σ is the neutron cross section, Φ is the neutron flux density, α is the isotopic abundance of the element, ε is the detection efficiency, j the quanta produced, and A is the atomic mass, respectively.

N_A, σ, Φ, α, ε, j, A and t are constants so the relation between N and G is:

$$G = p\,N + q \qquad (2)$$

where p and q are empirical constants determined experimentally. Eq. (2) represents the relation between

* E-mail: jingsw504@nenu.edu.cn

0236–5731/2004/USD 20.00

Akadémiai Kiadó, Budapest
Kluwer Academic Publishers, Dordrecht

G and *N* of a single element. In practice, there are a lot of elements in coal and they interfere each other. The multiple linearity regress formula[5] is employed to decrease the impact between one element and the others.

Design of the experimental setup

The experimental setup consisted of a pulsed neutron generator tube, a BGO detector system, a neutron monitor and a coal sample as shown in Fig. 1. The neutron yield was 10^8 ns^{-1}, the lifetime more than 2000 hours and the stability more than 0.5%. The detector system included a $\Phi 50\times 50$ mm^2 BGO detector main amplifier and 4096-channel MCA. The ZnS(Ag) detector was employed to monitor neutron flux. The $0.3\times 0.3\times 0.3$ m^3 coal sample box has been made of iron sheet.

The time sequence of neutron generator and gate circuit is shown in Fig. 2. Neutrons are emitted in the former 100 μs and cease in the rear 200 μs. Spectrum I (the overall spectrum of inelastic capture and activation) was obtained after 10 μs of neutron generating and lasted 80 μs. Spectrum II (the sum spectrum of capture and activation) was obtained after 210 μs of neutron generating and lasted 80 μs too. Spectra I and II was stored in the rear 2048- and former 2048-channels of the MCA, repsectively. To get a relatively pure inelastic spectrum a gate circuit mentioned above was designed, which can greatly improve the measurement precision.

In order to set up a satisfactory mathematic regression model with broad applicability and high precision, the standard samples made by the Coal Quality Supervise Inspection Center were used for the regression experiment. The coal samples from Shangyashan and Shulan city were made by standard method and sealed in the same iron boxes.

Results

Spectra I and II are shown in Fig. 3. Spectrum I is the overall spectrum of inelastic capture and activation and II is the sum spectrum of capture and activation. In spectrum I peaks of C, O and H are obvious. The other characteristic peaks of elements in coal are obvious in spectrum II. Carbon, hydrogen, oxygen and lead are necessary to calculate low caloric power ($Q_{\text{net ar}}$). The characteristic peaks and corresponding energy windows chosen in our experiment are given in Table 1.

I_{All} and I_{Cap} gotten in the experiment represent the sum counts of spectrum II and I, respectively, in order to decrease the impact of instability of neutron flux.

According to the data, spectra of standard coal samples the following multi linearity regression equations are obtained:

$$\begin{aligned} M_{\text{H}} &= 5.1682\, N_H/I_{Cap}-1.6404 \\ M_{\text{C}} &= [408.3868\,(N_{\text{C1}}-N_{\text{C2}})-865.6439 \\ &(N_{\text{O1}}-N_{\text{O2}})-23.1283\, N_{\text{Pb}}]/(I_{All}-I_{Cap})+11.3217 \\ M_{\text{O}} &= [1192.9342\,(N_{\text{O1}}-N_{\text{O2}})-225.0214 \\ &N_{\text{Pb}}]/(I_{All}-I_{Cap})-26.05442 \end{aligned} \quad (3)$$

where M_{H}, M_{C} and M_{O} are the mass of carbon, hydrogen and oxygen, respectively. Here, M_{O} and M_{H} include the mass of hydrogen and oxygen of water in coal. Accounted for the age of some coal or some similar coal mines, the ratio of hydrogen to carbon are in accordance with the same rule. The relationship between organic hydrogen (represented by *HY*) and carbon is given by:

$$HY = 0.0436M_{\text{C}}+0.2413 \quad (4)$$

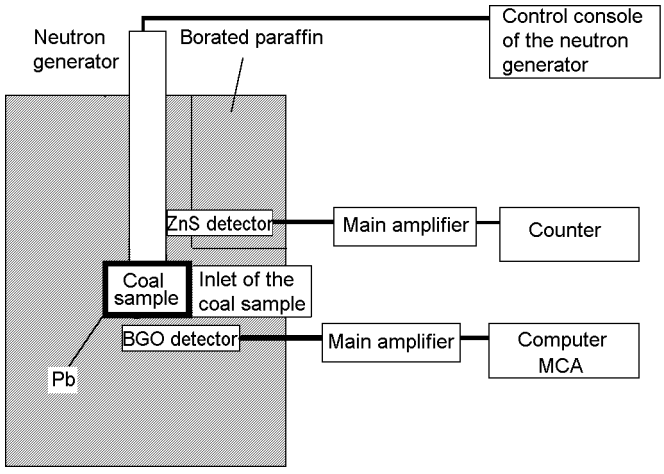

Fig. 1. The scheme of the measurement system

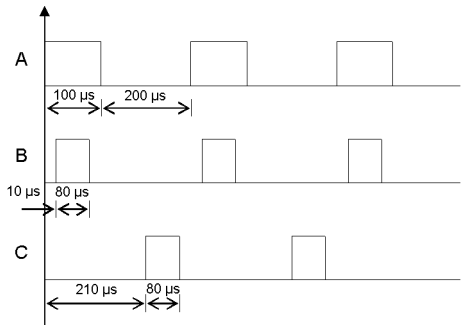

Fig. 2. Time structure of experimental conditions. Time structure of pulsed neutron generator (A); measurement time for spectrum I (B); measurement time for spectrum II (C)

The value of M_H–HY is the mass content of hydrogen in water. The mass of oxygen in water is known simultaneously. So the dispersion between M_O and oxygen mass in water is the mass of organic oxygen in coal (represented by HO). C_C, C_H and C_O (which represent the mass percent of organic C, H and O, respectively) are values of M_C, HY and HO divided by the total coal mass in the sample box.

According to the relation between the low caloric power of coal $Q_{net\ ar}$ and element percentage concentration in standard samples, the regression formula is:

$$Q_{net\ ar} = 0.4548C_C - 1.0198C_H + 0.1513C_O - 1.0924 \quad (5)$$

Using Eq (5), we analyzed the coal samples in Qitaihe, Hegang, Huolinhe, Zhusihua and Tumen coal mines and the results are shown in Table 2. Compared with chemical analysis, the main error of the system is 0.4 MJ/kg.

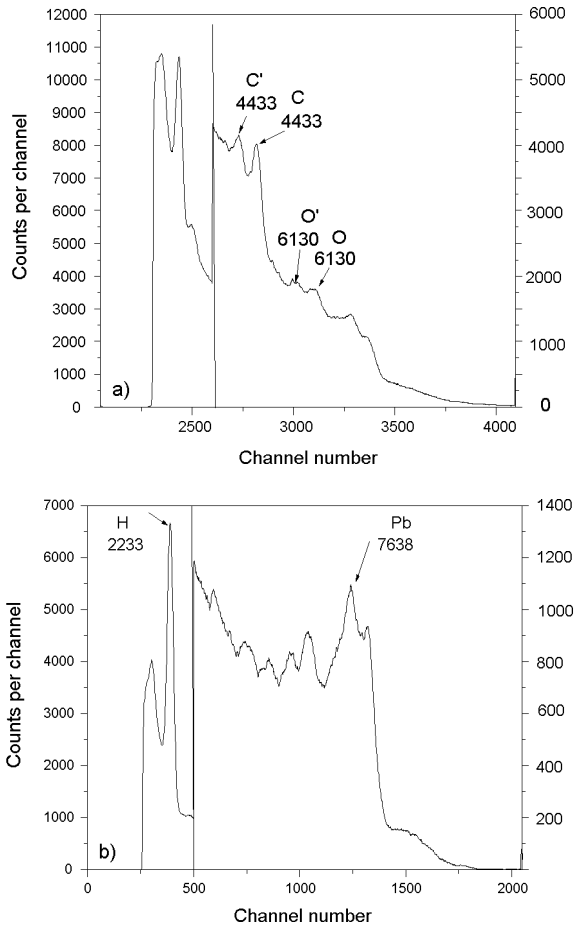

Fig. 3. Typical spectra of a coal sample. Overall spectrum of inelastic capture and activation (a), sum spectrum of capture and activation (b)

Table 1. Regions of interest to γ-ray spectrum

Element	Characteristic peak energy, MeV	Identification	Energy window of characteristic peak, MeV
C in spectrum I	4.43	N_{C1}	4.20–4.67
O in spectrum I	6.13	N_{O1}	5.89–6.36
Pb in spectrum I	7.36	N_{Pb}	6.78–7.59
C in spectrum II	4.43	N_{C2}	4.20–4.67
O in spectrum II	6.13	N_{O2}	5.89–6.36
H in spectrum II	2.23	N_H	2.00–2.47

Table 2. Coal sample analysis

Measurement date	Coal sample code	$Q_{net\ ar}$, MJ/kg	Value of assay, MJ/kg	Deviation, MJ/kg
2002.11.13	11120003	21.48 ± 0.22	21.88 ± 0.15	–0.40
2002.11.08	11030016	17.33 ± 0.30	17.28 ± 0.19	0.05
2002.11.09	11030020	16.39 ± 0.15	16.75 ± 0.27	–0.36
2002.11.13	11120008	22.90 ± 0.34	22.73 ± 0.17	0.17
2002.11.09	11090003a	13.27 ± 0.25	13.23 ± 0.22	0.04
2002.11.09	11080006a	13.38 ± 0.26	13.03 ± 0.29	0.35

Discussion and conclusions

A kind of coal sample can be detected in 15 minutes by the pulsed neutron method, which is faster than conventional chemical analysis. The error of low caloric power $Q_{net\ ar}$ of coal between the pulsed neutron method and conventional chemical analysis is below 0.4 MJ/kg, which meets the coal industry demand and can direct the industrial production. However, we have noted that the results are greatly dependent upon the quality of the calibration. We should classify coal species of different coal mines and set up corresponding mathematic models so that the system can meet more different coal species and coal mines. At the same time, the temperature effect of the BGO detector and the flux instability of neutron generator are influencing the detection precision greatly, a problem which should be solved in the future.

*

This research was supported by the innovation fund for small technology-based firms under Contract No. 99C26212210085 and also by the Science Foundation for Young Teachers of Northeast Normal University.

References

1. M. R. WIRNAKD, C. G. CLAYTON, J. Appl. Radiation Isotopes, 34 (1983) 7.
2. C. G. CLAYTON, M. R. WIRNAKD, J. Appl. Radiation Isotopes, 34 (1983) 83.
3. C. G. CLAYTON, M. R. WIRNAKD, J. Appl. Radiation Isotopes, 34 (1983) 3.
4. G. VOURVOPOULOS, Nucl. Instr. Meth. Phys. Res., B56/57 (1991) 917.
5. M. BLAAUW, R. F. FLEMING, R. KEYSER, J. Radioanal. Nucl. Chem., 248 (2001) 309.