Improvement of the determination of hydrogen content in a multicomponent sample by D–T generator *

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Abstract If a D–T generator is used as a neutron source to simultaneously measure the content of carbon, hydrogen and oxygen in a multicomponent sample by NIPGA (Neutron Induced Prompt Gamma-ray Analysis), the 14 MeV neutron flux can be regarded as a constant value. The relationship between the production of the hydrogen characteristic gamma-rays and its content is nonlinear. In this paper, we use MCNP (Monte Carlo N-Particle Transport code) to simulate the relationship and analyze it. In practical measurement of the characteristic gamma-ray, it's impossible to get the net count. Therefore, we use the experiment to obtain the relationship between the hydrogen content and the total count of its characteristic gamma-rays. If we use the relationship combined with the simulation result to calculate the hydrogen content, the metrical precision can be much increased. The deviation of hydrogen content between NIPGA and chemical analysis is less than 0.25%, which meets the requirement of coal industry.

Key words NIPGA, hydrogen content, D–T neutron generator, MCNP, nonlinear

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1 Introduction

In order to increase the economic profit, the enterprises which use, for example, coal as fuel need to quickly and simultaneously measure the contents of hydrogen, carbon and oxygen in coal. The conventional chemical method needs a long time to be carried out, for instance, ten hours, therefore it cannot direct the industrial product on time [1]. NIPGA is a fast analysis technique, which can analyze a multicomponent sample in 15 min. As a result, it is widely used to analyze the contents of the multicomponent sample, for example, coal.

In NIPGA, the prompt characteristic γ-rays which are induced by inelastic scattering are used to analyze the contents of carbon and oxygen, but, determining the hydrogen content needs the γ-rays induced by thermal neutron capture. In analyzing the coal properties by NIPGA, the contents of carbon, hydrogen and oxygen have to be measured simultaneously. Therefore, a D–T generator is often used as a neutron source [2, 3]. The 14MeV neutron flux can be regarded as a constant value, but the thermal neutron flux is relevant to the coal component. The relationship between hydrogen and its characteristic intensity is nonlinear. But in many papers a linear relationship is still to be taken [4, 5]. It will increase the measurement errors and cannot meet the requirement of enterprise. Therefore, the NIPGA would be unpromising in analyzing coal properties. We used MCNP to simulate the relationship with a constant fast flux after analyzing the relationship between the production of the hydrogen characteristic gamma-rays and its content in sample. Then，the experiment was used to validate and amend the relationship.

2 Principle

If a hydrogen nucleus captures a thermal neutron,

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it will emit a γ-ray at 2.22 MeV. In quantitative analysis, the relationship between the hydrogen content and the net count of its characteristic γ-rays is as follows:

$$N=\frac{GN_{\mathrm{L}}\varphi\sigma\varepsilon j\alpha t}{A}\ . \tag{1}$$

where, N is the net count of the characteristic γ-ray of hydrogen (in measurement period t), G is the hydrogen content in coal sample, N_{L} is the Loschmidt constant, φ is the thermal neutron flux density, σ is the thermal neutron capture cross-section of hydrogen, ε is the detection efficiency of γ-ray detector, j is the quanta produced, α is the isotopic abundance of the element, and A is the atomic mass.

Obviously, the parameters of N_{L}, σ, j, α, t and A are all constants. For a γ-ray detector used in the measurements, its ε is invariable. In practical application, it's impossible to get the net count of the characteristic γ-ray. If the total counts and the background counts are expressed as N_{t} and b respectively,

$$N_{\mathrm{t}}=N+b=p\varphi G+b\ . \tag{2}$$

where, $p=N_{\mathrm{L}}\sigma\varepsilon j\alpha t/A$.

Once the D–T neutron generator is selected, the fast neutron flux can be regarded as a constant value. The 14 MeV neutrons have to collide with nucleus in the sample to decrease their energies and become thermal neutrons. Then, the hydrogen nucleus may capture them and emit characteristic γ-rays at one time. Because the ability to moderate neutrons is different, the thermal neutron flux is relevant to the contents of all the elements in coal. In the process of moderating neutrons, the smaller the atomic mass is, the better the ability to moderate neutrons is. The mass number of carbon in coal is the mainly smallest one except hydrogen. But its ability to moderate neutrons is much lower than the hydrogen's. Although the mass percentage of hydrogen in coal is about 5%, the mole percent is about 50%. As a result, the thermal neutron flux is closely relevant to the content of hydrogen in coal.

3 Simulation of the relationship between *G* and *N*

The MCNP is employed to simulate the relationship between G and N. The scheme of the simulation system is shown in Fig. 1. D–T neutron generator regarded as a neutron point source is placed on the coordinate origin. In order to compare the simulation result with the experimental one, the coal sample is placed in a cube whose length, width and height are 15 cm, 20 cm and 20 cm, respectively. The gamma-ray detector is a cylinder of 3.8 cm in radius and 7.6 cm in height. The 14 MeV neutron flux is 10^8 n/s, and the measure time is 900 s. The simulation result of N as a function of G is shown in Fig. 2. As can be seen from Fig. 2, N is proportional to the third power of G.

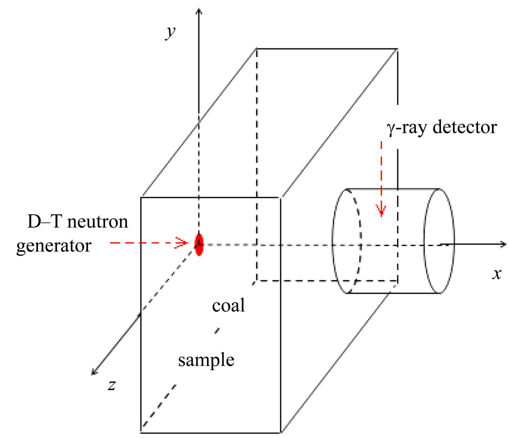

Fig. 1. Scheme of simulation system.

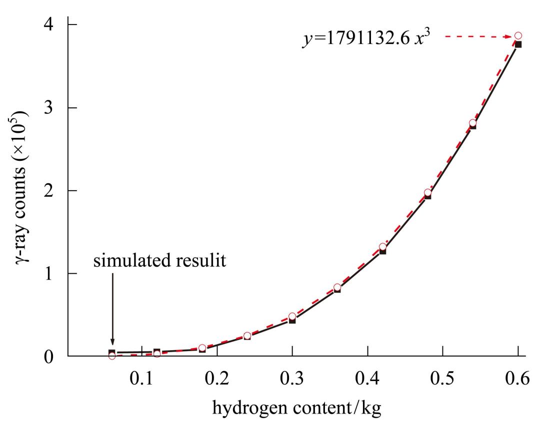

Fig. 2. Simulated result of N as a function of G.

The ability of medium to moderate neutrons is proportional to the nucleus number. Because the ability of hydrogen nuclear to moderate neutrons is much higher than the other atom's, the thermal neutron flux is approximatively proportional to the hydrogen content in coal. Of course it's also proportional to the fast neutron flux:

$$\varphi=k_1\varphi_{\mathrm{fast}}G. \tag{3}$$

where, φ_{fast} is the fast neutron flux.

The probability of thermal neutron capture is approximatively proportional to the second power of nucleus number. Because the number of other nucleus which can capture thermal neutrons is much less than the hydrogen's, the number of the thermal neutron captured by nucleus is approximatively proportional to the second power of the hydrogen atom's number. The number of the characteristic γ-rays is equal to the number of the thermal neutrons captured by hydrogen, so

$$N=k_2G^2\varphi=k_2G^2(k_1\varphi_{\rm fast}G). \tag{4}$$

If the fast neutron flux is invariable,

$$N=k_3G^3. \tag{5}$$

Considering the background of the γ ray,

$$G=\left(\frac{N_{\rm t}-b}{k_3}\right)^{1/3}=k(N_{\rm t}-b)^{1/3}. \tag{6}$$

where, $N_{\rm t}$ is the total number of the γ rays detected in the peak region, k is a coefficient determined by the coal sample, the fast neutron flux, the detection efficiency of γ-ray detector and measurement time.

4 Experimental measurements

The experimental setup consists of a D–T neutron generator provided by Northeast Normal University, a γ-ray detector system, and a sample, as shown in Fig. 3. The neutron yield is 10^8 n/s, the lifetime is more than 2000 h and the stability is more than 0.5%. The γ-ray detector system includes a BGO (Bismuth Germanate) crystal whose radius is 3.8 cm and height is 7.6 cm, a main amplifier, a 4096-channel MCA (Multi-Channel Analyzer) and a computer. The BGO detector and MCA were offered by the Shanghai Silicate Institute and the Shanghai Atomic Nuclear Institute. The coal sample is placed in a 0.2 mm×0.2 mm×0.15 mm box. A typical spectrum of γ-rays is shown in Fig. 4.

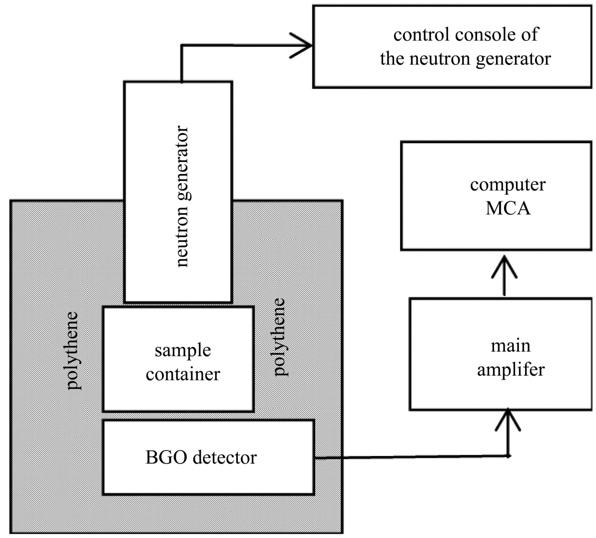

Fig. 3. Scheme of the experimental setup.

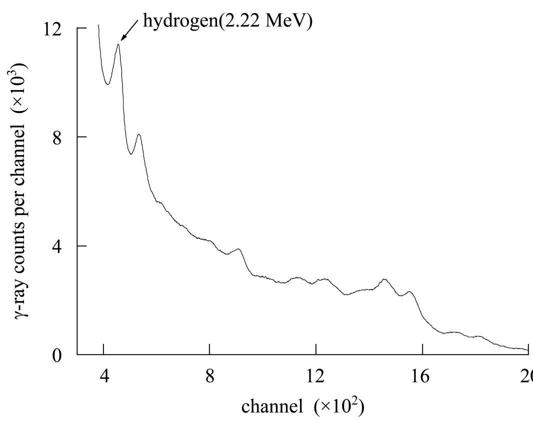

Fig. 4. Spectrum obtained of γ-ray.

Table 1. Measurement results from the standard samples.

sample	M/kg	$H_{\rm c}$(%)	$N_{\rm t}$	$H_{\rm n}$(%)	D(%)	$H_{\rm L}$(%)	$D_{\rm L}$(%)
1	6.27	1.82	231913	1.78±0.15	−0.04	2.86	1.04
2	6.01	2.36	231982	2.38±0.16	0.02	3.06	0.70
3	6.09	2.91	232093	2.88±0.12	−0.03	3.14	0.23
4	6.06	3.59	232326	3.63±0.14	0.04	3.41	−0.18
5	5.94	4.47	232715	4.52±0.11	0.05	3.91	−0.56
6	6.13	5.16	233227	5.11±0.12	−0.05	4.35	−0.81
7	6.11	6.08	234087	6.03±0.14	−0.05	5.30	−0.78
8	5.97	6.84	234905	6.85±0.16	0.01	6.33	−0.51
9	5.94	7.56	236015	7.63±0.13	0.07	7.60	0.04
10	6.07	8.32	237553	8.29±0.10	−0.03	9.12	0.80

The standard samples were made by the Coal Quality Supervision Inspection Center of China. Their data are shown in Table 1. M is the mass of the coal sample, $H_{\rm c}$ is the mass percentage of hydrogen measured by chemical method, $N_{\rm t}$ is the total γ-ray counts of hydrogen which includes the background counts. According to the data measured for the standard samples, the mass percentage of hydrogen is

$$H_{\mathrm{n}} = \frac{0.02817 \times (N_{\mathrm{t}} - 231861)^{1/3}}{M}. \quad (7)$$

Each standard sample was measured ten times. The results are shown in Table 1. D is the difference of H_{n} and H_{c}.

Table 2. Measurement results from different mines.

coal sample	H_{c}(%)	H_{n}(%)	D(%)
20090810a2	4.39	4.54±0.23	0.15
20090814a5	2.35	2.53±0.17	0.18
20090912a3	6.19	6.06±0.21	−0.13
20090912a6	5.22	5.41±0.17	0.19
20090913a4	3.47	3.37±0.18	−0.10
200901015a1	3.16	3.32±0.17	0.16
20091017a6	4.22	4.07±0.19	−0.15
20091112a2	6.04	6.24±0.13	0.20
20091114a4	5.37	5.20±0.15	−0.17
20091116a3	4.92	4.77±0.16	−0.15

Each coal sample from Qitaihe Coal Mine, Hegang Coal Mine, Huolinhe Coal Mine, and Tumen Coal Mine in China was measured for ten times. The data measured for the samples were analyzed by using Eq. (7). The results are shown in Table 2. The maximal deviation is less than 0.25%, which sufficiently satisfies the requirement of coal industry [1].

If using the linear dependence of N on G as Refs. [4, 5], according to the data of the standard samples, the mass percentage of hydrogen is

$$H_{\mathrm{L}} = \frac{6.645 \times 10^{-5} \times N_{\mathrm{H}} - 15.23}{M}. \quad (8)$$

The results are shown in Table 1. D_{L} is the difference of H_{L} and H_{c}. There are only three standard samples whose deviations are less than 0.25%.

5 Conclusions

In this paper, the relation of the net counts of the hydrogen characteristic gamma-ray to its content in a sample was simulated for NIPGA by using MCNP code. The result shows that the net count is dependent on the third power of its content in the sample. The experiment was used to validate and amend the relation. The deviations of hydrogen content between NIPGA and the chemical method were all less than 0.25%, which is much improved compared with the linear relation and sufficiently satisfies the requirement of coal industry.

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