Progress of neutron induced prompt gamma analysis technique in 1988~2003

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Abstract This paper describes new development of the neutron induced prompt gamma-ray analysis (NIPGA) technology in 1988~2003. The pulse fast-thermal neutron activation analysis method, which utilized the inelastic reaction and capture reaction jointly, was employed to measure the elemental contents more efficiently. Lifetime of the neutron generator was more than 10000 h and the performance of detector and MCA reached a high level. At the same time, Monte Carlo library least-square method was used to solve the nonlinearity problem in the NIPGA.

Keywords Inelastic scattering, Thermal neutron capture, Neutron induced prompt gamma-ray, On-line element analysis, Monte Carlo simulation

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1 General situation

In the late of 1980s and the early of 1990s, researchers found that there were still difficulties in analyzing some light elements (such as C and O) by NIPGA and wanted to employ neutron inelastic reaction (n, n' γ) to measure them, and to solve some unsolved problems by NIPGA.[1-3] In 1987, Gozani firstly employed the prompt inelastic neutron activation analysis (PINAA) to measure elemental content in the interrogated materials[4] and developed a new explosive detection system named SPEDS in 2003 with the least measurable mass of 100g.[5] Thorpe analyzed the elements in coal in 1988 by using neutron generator, and concluded that the 14 MeV neutron generator was only useful for C and O measurement, and the determination of other elements in coal still relied on the thermal neutron capture reaction.[6]

Chen Bo-Xian *et al*. used the Am-Be neutron source to analyze the elements in coal in 1996. They demonstrated that the elements C, O and Si could be measured by [n, n' γ] reaction, which was the supplement of the [n, γ] reaction.[7] Liu Yu-Ren and Hu Shu-Zhi expressed the same opinion.[8]

In 1998 Vourvopoulos, Dep and Belbot measured the elements in coal on-line by using the pulse neutron generator and BGO detector. They believed that the PFTNA (pulse fast-thermal neutron activation) was a promising method because the detection precision for S and C was 0.05wt% and 1wt%, respectively.[9]

Belbot and Vourvopoulos of Western Kentucky University developed a prototype of coal element analysis in 1999 by employing the pulse neutron source, and the prototype was applied on in-situ activation analysis. The analysis error of elements O, C and S was 2%, 0.6% and 0.18%, respectively. The BGO detector and D-T pulsed neutron generator were utilized in the prototype, which was used to detect explosives by comparing the C/O and C/N ratio.[10]

Subsequently, Letourneur *et al*. of SODERN Company in France analyzed the bauxite by PFTNA. Their results showed that 99% of the raw material could be analyzed by using the sealed pulsed neutron tube, so the PFTNA technology could be applied to the inspection of aluminum production. In 2002, they adopted the same method to analyze the elements in coal. It was reported that Lim *et al*. developed a new on-belt element analyzer of cement raw material by

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using the PFTNA system in 2001. The detection error after 10 minutes' counts for CaO, SiO_2, Al_2O_3 and Fe_2O_3 was 0.49 wt%, 0.52 wt%, 0.38 wt% and 0.23 wt%, respectively.[11]

In China, the Nanjing Dalu Zhongdian Company and Northeast Normal University developed the same work and got satisfying results in 2001 and 2002,[12] respectively.

2 Physical method

In the 1980s to 1990s, the Monte Carlo simulation was used to design the experimental facility and to evaluate the physical method. The first thing is to understand the thermal and fast neutron distribution in the material. According to the simulation result, the optimum detection volume of the material and the distance between the source and the detector can be decided. At the same time, with nuclear reaction data of the related elements, the measurement precision can be estimated. But the result is approximate. The theoretical analysis and experimental study are introduced as follows.

2.1 Neutron distribution in coal by the Monte Carlo simulation

Oliveira and Salgado studied the thermal neutron distribution of ^{241}Am-Be source in coal using the Monte Carlo code[13] in 1990. The result showed that the sample thickness should be more than 25 cm, because more photons appeared in 25 cm sphere close to the detector.

In 1991, Oliveira and Salgado also employed the Monte Carlo code to study the fast neutron distribution of ^{241}Am-Be source in coal.[14] The result also showed that there were more photons in 15cm sphere close to the source.

In 1993 Oliveira *et al*. studied the thermal neutron distribution of ^{252}Cf with moderator by using the Monte Carlo code.[15, 16] The results showed that the gamma counts in the detector did not vary with V_H, the volume content of H, when the sample thickness was less than 20 cm and the radius of moderator was more than 7.5 cm.

In 1998 Dep *et al*. employed the MCNP (Monte Carlo Neutron Photon) to design the NIPGA[9] including the sample volume and the size of moderator and reflector. The same work was developed in China, which aimed at the 14 MeV fast neutrons.

2.2 Experimental study of element content in coal using 14 MeV neutron generator

We once mentioned that Arthur *et al*.[7] analyzed the elements in coal using the neutron generator. They believed that it was a long distance before the 14 MeV neutrons were slowed down to become thermal neutrons, and thus resulting in a loss of resolution. If the neutron generator produces the 2.5 MeV neutrons, the resolution would increase dramatically. The authors compared the 14 MeV neutron generator with 2.4 MeV ^{252}Cf neutron source. The result showed that it is disadvantageous for [n, γ] reaction by using the 14 MeV neutron generator but it is good for [n, n' γ] reaction to measure C and O.

In 1995 Womble *et al*.[17] employed D-T neutron generator with a 4π yield of 4×10^8 s^{-1} and a lead reflector in their experiment and obtained a good result. Then, the Western Kentucky University group developed a prototype of pulsed neutron coal analyzer (mentioned above).

2.3 Neutron moderator, reflector and shield material

With the development of industry, new products of neutron moderator, reflector and shield material have appeared.[8] For example, the zirconium deuterated and colophony with heavy water is used for moderated material. The WEP662P colophony with 70%~80% heavy water has good character for machining. The perfect material for neutron shield is polythene with boron carbonized.

2.4 Measurement method

Based on the result mentioned above, we can conclude that the NIPGA technology has entered a new stage of the mix of [n, n' γ] and [n, γ] reaction. Lindstrom *et al*. employed thermal neutrons to analyze the small volume samples in 1980[18] and they believed that the sample volume could be very small if the thermal neutron flux was high enough (the neutron flux they employed was 2×10^8 s^{-1}·cm^{-2}). This result corrected the wrong cognition that NIPGA could only be used to analyze large volume sample. In 2001 Lim *et al*. employed the technique mentioned above to develop an on-line analyzer for cement raw material. They utilized the dual-source and dual-detector to decrease the error caused by the distribution asymmetry of the sample on belt.[11]

3 Neutron source and measurement facilities

3.1 Neutron source

The neutron source for NIPGA is ^{241}Am-Be or neutron generator. A perfect neutron source is the neutron generator with long lifetime. According to the introduction of EADS Company (Germany) in 2001, the lifetime of Fusionstar FS-NG1, one of its products, is over 10000 h, with a neutron yield of the D-T generator equal to 1.6×10^8 s^{-1}. The stability of the neutron generator produced in China is about 0.5%.

3.2 Measurement facilities

NaI, BGO and HPGe detectors were used in neutron activation analysis from the 1970s. For fast and thermal neutron on-line analysis, efficiency of BGO detectors is higher than any other detectors, but its resolution is low. On the contrary, resolution of HPGe is high and the relative efficiency is more than 100%, but it is inconvenient to on-the-spot application because liquid nitrogen is needed and the resolution may be ruined by neutron damage. The use of NaI detector is under consideration in recent 2 years. For example, Gardner *et al*. studied the NaI spectrum of NIPGA in detail.[19] In 2000, Ghanem in Alexandria University of Egypt simulated the characteristic response of NaI detector by using the Monte Carlo code.[20] In 2001 Prettyman *et al*. published the research result on TeZnCd deterctor.[21] This kind of TeZnCd is better than NaI (Tl) on resolution and can detect the gamma rays more accurately.

As for the MCA, Canberra and ORTEC Company in USA developed DSP-2000 and DSPEC gamma spectrometer, respectively. DSP-2000 is a digital analyzer with high stability, precision and repetition rate. In the application of high throughput rate, DSP-2000 is the best choice because its throughput rate reaches 100,000 per second. Both DSPEC and DSP-2000 can be employed in the NIPGA application. It was proved that the analysis result was better when the digital MCA was employed.[3] In 2002 Beijing International Nuclear Industry Exhibition, ORTEC exhibited its new MCA named Digidart with a maximum system throughput of more than 100,000 cps and 16384 channels though its volume is small.

4 Data management-analysis software

There are different features with the data management of NIPGA in different periods. The earliest methods is the comparison with the standard material, the second one is the regression, and the recent one is the Monte Carlo simulation.

The first method can not be employed in on-the-spot applications, therefore the regression method is often utilized instead. The latter method is based on the relation between the element contents and the area of the characteristic gamma rays. To obtain the correlation coefficient, many runs of experiments are needed. But times of experiment can be decreased by using the regression test design,[22] and the regression analysis method can be improved by using the “partial least-square regression”.[23]

In 1991 Shyu Charur-Ming developed a data management software for NIPGA by using the Monte Carlo library least-square simulation.[24] The following functions were included:

a) Simulate the spectrum response of the known content sample with the Monte Carlo method;

b) Take track of the library spectrum response of each element;

c) Get the unknown samples’ content with linear library least-square;

d) Improve the precision with the overlapping method.

In 1998 Gehrke *et al*. evaluated the Monte Carlo library least-square method and believed that it could solve the nonlinearity problem and expand its application area.[25] The recent work of Gardner was to apply the MCNP, and to develop the CEARPGA (Center for Engineering Applications of Radioisotopes Prompt Gamma Analysis) in the end. The CEARPGA included a lot of useful contents, but MCNP code was still utilized by a lot of researchers.[26-29]

The parameters such as calorific value, moisture, ash and volatile contents can be obtained from the calculation according to the empirical formula.[17]

5 Conclusions

According to the introduction above, we know that the NIPGA technique has reached the level of the mix of [n, n'γ] and [n, γ] reaction. The neutron generator with a lifetime of more than 10,000 h gradually displace the radioactive neutron sources. At the same time, yield of the neutron generators reaches more than $1\times10^{8}\ s^{-1}$, and the stability is better than 0.5%. The NaI, BGO and HPGe detectors and the digital MCA have been employed in this application, and software is changing from MCNP to CEARPGA[30] mainly for NIPGA application. Furthermore, the neutron moderator, reflector and shield material have developed in the past 10 years. All in all, it is the good time for NIPGA technique to be applied in industry.

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