



The Review of Prompt Gamma Activation Analysis

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Abstract

Prompt gamma activation analysis (PGAA) has the characteristics of high sensitivity and non-destructive analysis. At the same time, compared with neutron activation analysis, prompt gamma activation analysis requires less neutron flux and causes lower induced radioactivity to the sample. After a period of time, the radioactivity of the sample will be reduced to a natural level and can be used for other studies. Therefore, PGAA is also a suitable choice for some precious samples. PGAA has been applied in various fields, especially for the determination of light elements such as hydrogen, boron and other elements in large samples. At the same time, it also has non-destructive testing properties. It can be applied in various fields and has broad development prospects. Therefore, this paper summarizes the facilities and research profiles of PGAA system at home and abroad and its application profiles, understands the current development status of PGAA system, and lays a certain foundation for future research. The emphasis of this review is on laboratory measurements based on reactor neutron beam. The application of on-site prompt gamma activation analysis is not discussed in this paper. It is found that although PGAA can detect most of the elements on the periodic table, the detection sensitivity is lower than that of neutron activation analysis. The detection ability of the system can be improved by modifying the experimental facilities, such as shielding the background and using the anti-Kang spectrometer.

Subject Areas

Nuclear Physics

Keywords

PGAA, Basic Principle, Analytical Methods, Applications

1. Introduction

PGAA is a non-destructive and highly sensitive measurement method for the elements in the material by using the reaction of capture, elastic and inelastic scattering after neutron beam bombardment of the sample. It can measure some elements quickly and accurately, and has many advantages, such as simultaneous analysis of multiple elements and adaptation to the current international “hydrogen economy” idea.

At present, many experimental platforms based on PGAA technology have been established at home and abroad, and a wide range of application research has been carried out. For example, the University of Texas in the United States, the University of Mainz in Germany, the Korea Institute of Atomic Energy and other institutions have studied the application of PGAA technology in related fields. This paper aims to make a comprehensive overview of the current research at home and abroad, so as to better understand the existing research progress of the technology and facilitate the determination of the next research direction.

2. PGAA Principle

2.1. Basic Principle

Prompt gamma activation analysis, collectively referred to as PGAA (Prompt Gamma Activation Analysis), uses neutron beams to bombard the sample. The nucleus of the sample captures neutrons and generates a composite nucleus in the excited state. The composite nucleus deactivates and releases gamma rays. Different nuclides correspond to gamma rays of different energies. These characteristic gamma rays are analyzed using a high-resolution gamma spectrometer. According to the intensity of the characteristic peaks, the composition and content of these nuclides can be determined to achieve qualitative and quantitative analysis [1] [2] [3].

Figure 1 shows the difference between neutron activation analysis (NAA) and prompt gamma activation analysis. According to the different sources of characteristic gamma rays, NAA analyzes the characteristic gamma rays produced by the decay of composite nuclei, and prompt gamma activation analysis analyzes the characteristic gamma rays produced by the deexcitation of composite nuclei.

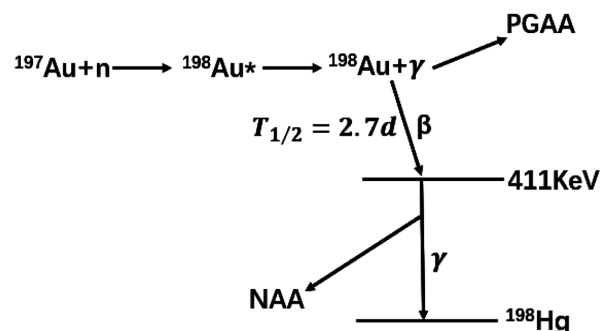


Figure 1. Principle of NAA and PGAA.

2.2. PGAA Analysis Method

PGAA analysis method is divided into k_0 method, relative method and calibration curve method. The k_0 value can be measured or calculated by experiment, where the k_0 value formula is as follows [4] [5] [6] [7] [8]:

$$k_{0,c}(x) = \frac{N_{\gamma,x}/\varepsilon_{\gamma,x} \cdot m_c}{N_{\gamma,c}/\varepsilon_{\gamma,c} \cdot m_x} \quad (1)$$

where N_γ is the net peak area of gamma ray, ε_γ is the full energy peak detection efficiency, and m is the mass of the element x to be measured relative to the comparator c . k_0 method to measure element content ω_x formula is as follows:

$$\omega_x = \frac{\varepsilon A_{sp,x}}{A_{sp,c} \cdot k_{0,c}(x) \cdot \varepsilon} \quad (2)$$

$A_{sp,x}$, $A_{sp,s}$ represent the characteristic peak area of the sample to be tested and the standard sample respectively.

The relative formula is as follows:

$$\frac{\omega_x}{\omega_s} = \frac{A_{sp,x}}{A_{sp,s}} \quad (3)$$

where ω_x and ω_s represent the element content in the sample to be tested and the element content in the standard sample, respectively. Analysis using the relative method requires the production of a standard sample with the same geometric conditions as the sample to be tested, and the measurement conditions are the same as the sample to be tested.

Calibration curve method [9], calibration curve method is improved for the relative method, compared to the relative method of measurement accuracy. It is mainly to measure a series of standard samples with known element content, establish a relationship curve between characteristic peak intensity and element content, and then use the calibration curve to obtain the element content.

3. Domestic and Foreign Facilities and Research Overview

3.1. CARR PGAA System

In the 1990s, the China Institute of Atomic Energy built a PGAA experimental platform based on the heavy water reactor (101 reactor) and studied the high-energy γ -ray wide-energy zone calibration technology and the single comparator k_0 method. The submarine manganese nodule reference material was also analyzed, and the analysis results were consistent with the standard values. However, the neutron beam of the 101 reactor is not ideal, and there are problems of low neutron flux and high background. The China Advanced Research Reactor (CARR) newly built by China Institute of Atomic Energy (CIAE) can provide high-quality neutron beam, which is characterized by large quality factor and large effective neutron utilization space. The neutron flux can reach $1.2 \times 10^{15} \text{ cm}^{-2} \cdot \text{s}^{-1}$ at 60 MW full power operation. Activation Analysis Group of China Institute of Atomic Energy will build short-lived nuclide Neutron Activation

Analysis (NAA) system, Prompt Gamma Thermal Neutron and Cold Neutron Activation Analysis (PGAA) system and Cold Neutron Depth Profile Analysis Invert (NDP) system based on CARR to provide advanced experimental platform for nuclear analysis in China.

At present, some work has been carried out on the PGAA system of CARR reactor, including the design and simulation calculation of the shielding system of PGAA, and the geometric structure and size of the shielding system have been determined [10], which provides the basis for the construction of the thermal neutron prompt gamma activation analysis system. Yun Weixu [11] designed and debugged the relevant parameters of the cold neutron prompt gamma activation analysis (CNPAA) measurement device, and built the first set of cold neutron prompt gamma activation analysis system in China. Sun Hongchao [12] carried out the neutron self-shielding correction under the condition of reactor beam for the first time in China, and established the prompt gamma activation analysis system based on beam regulation technology for the first time in China. Xiao Caijin and Yao Yonggang [13] carried out the experiment for the first time in the cold neutron prompt gamma activation analysis system of CARR reactor, and obtained the cold neutron prompt gamma spectrum of NH_4Cl . At the same time, the energy calibration of the detector in the wide energy range of 0.1 KeV to 8 MeV was carried out.

3.2. Foreign PGAA System

With the development of high purity germanium detector and fast electronics, prompt gamma activation analysis has been developed rapidly. At present, more than thirty laboratories abroad have carried out prompt gamma activation analysis. **Table 1** lists the PGAA system built or planned after 1990.

4. Application of PGAA

4.1. Materials Science

PGAA has the advantages of high sensitivity and non-destructive testing for the determination of hydrogen. The determination of hydrogen is realized by the 2223.25 KeV gamma ray emitted by the $1\text{H} (n, \gamma) 2\text{H}$ reaction. A lot of research has been carried out on PGAA in materials, especially hydrogen measurement. The University of Texas [29] used the PGAA device to determine the hydrogen content of metal oxides, and proposed some error sources, including H in the environment, changes in neutron flux, spectral interference, sample self-absorption and neutron scattering. Danyal Turkoglu [30] studied titanium alloys containing trace hydrogen. The feasibility of using PGAA technology to determine trace hydrogen in metal samples was verified by measuring the sample packaging material embryo, prepared standard samples, titanium alloy SRMs samples and degassing titanium alloys. For materials containing high hydrogen [31], PGAA can detect the change of hydrogen content in the material, which proves the feasibility of this technology for measuring the water absorption of composite materials.

Table 1. PGAA systems built or planned after 1990.

location	neutron flux (cm ⁻² .s ⁻¹)	Year of completion	reference
CRN, Strasbourg (cold)	1 × 10 ⁶	1990	
DINR, Vietnam (thermal)	5 × 10 ⁶	1992	[14]
BNC, Budapest, Hungary (guide)	2 × 10 ⁶	1993	[15]
MIT, Cambridge, MA, USA (diffracted)	6 × 10 ⁶	1993	[16]
NBSR, Gaithersburg, MD, USA (cold)	1.5 × 10 ⁸	1993	[17]
JAERI, Tokai, Japan (cold and thermal)	1.4 × 10 ⁸ , 2.4 × 10 ⁷	1993	[18]
NBSR, Gaithersburg, USA (cold guide)	8 × 10 ⁸	1996	[17]
Univ. of Texas, Austin, USA (cold guide)	5 × 10 ⁷	1997	[19]
SINQ, Villigen, Switzerland (spallation, cold)	7 × 10 ⁷	1997	
Rez, Czech Republic	3 × 10 ⁶	2000	
Dhruva, India (thermal guide)	1.4 × 10 ⁷	2001	[20]
HANARO, Korea	8 × 10 ⁷	2002	[21]
RA-3, Argentine	10 ⁹	2003	[22]
FRM II, Garching, Germany	6 × 10 ⁹	2007	[23]
MEPhI, Russia		2009	[24]
Univ. of Oregon, Corvallis, USA	2.81 × 10 ⁷	2010	[25]
Univ. of Ohio, Columbus, USA	1.27 × 10 ⁷	2012	[26]
TRIGA IPR-R1, Brazil	10 ⁶		[27]
TRIGA Mark II, Moroccan		under construction	[28]

The PGAA device of the HANARO experimental reactor of the Korea Institute of Atomic Energy [16] measured the hydrogen in coal and metal samples. It was found that when the hydrogen concentration was less than 1 mg/kg and the sample mass was less than a few mg, the environment had a great influence on the hydrogen determination results. PGAA technology is also used to study hydrogen storage materials [32], for the study of the kinetics of hydrogenation reactions in metals [33] has important applications, but also for the study of lithium battery cathode materials [34] also has applications.

An important application of prompt gamma activation analysis technology is to study the hydrogen in zirconium alloys [35] [36] [37], which is of great significance in the study of hydrogen adsorption and corrosion resistance of zirconium alloys. The hydrogen absorption coefficient of zirconium alloys can be measured by prompt gamma activation analysis and vacuum thermal extraction technology. The hydrogen concentration of 5 wt. ppm in the alloy can be detected on the prompt gamma activation analysis device of NIST in the United States. The error is about ±1 wt. ppm, which is lower than that of traditional detection technology such as vacuum thermal extraction method, and will not affect the hydrogenation process and corrosion process of zirconium alloy samples.

4.2. Standard Reference Samples

The standard reference sample is an important strategic material of the country. In view of the fixed value of the element content in the standard reference sample, the prompt gamma activation analysis technology can also play an important role. Paul [38] used the combination of cold neutron prompt gamma activation analysis and thermal neutron prompt gamma activation analysis to determine the mass ratio of S/H and the mass fraction of H in the fuel SRM reference material. The analysis results of three SRM reference materials show that this method can identify sulfur with mass fraction $\geq 1\%$, and the relative error is about 0.1%. Becker [39] *et al.* analyzed the SRM 1573a tomato leaf reference material, providing more than 30 elements such as H, B, N, and K by combining PGAA with INAA and RNAA. Vogt [40] *et al.* determined the NBS 1633 A coal ash standard sample through PGAA, and determined ten elements. The results were consistent with the certification results of the National Bureau of Statistics (NBS). Sudarshan [41] combined internal standard method with PGAA technique to analyze three different IAEA CRM standard reference samples. Anderson [42] measured boron in more than thirty kinds of food and biological standard samples. The detection limit of the device is between 0.3 and 0.8 $\mu\text{g/g}$. For samples with boron content above 5 $\mu\text{g/g}$, the accuracy is about 1%. Maria [43] developed an innovative sugarcane leaf reference material, using a combination of PGAA and NAA methods to analyze sugarcane leaf reference materials. PGAA allows the determination of B, Cl, Fe, K, Mn, N and Si. Similar mass fraction values were obtained for those elements determined by nuclear technology in sugarcane leaf reference materials and SRM1570 and SRM1572, and the standard deviation of PGAA for element detection results was below 5%.

4.3. Biological Samples

PGAA can also be used to determine elements in biological samples such as boron, cadmium, etc., which are critical for studies such as boron neutron capture therapy (BNCT). The University of Mainz in Germany [44] compared PGAA in blood samples and healthy samples of four patients with colorectal cancer liver metastasis. It is believed that the pharmacokinetics becomes complicated during BNCT treatment, so it is necessary to use PGAA to measure boron content in blood. C. L. Schütz *et al.* [45] compared the results of inductively coupled plasma mass spectrometry, neutron capture photography, and prompt gamma activation analysis of boron in biological samples for the first time. The difference between the measurement results of boron in blood by ICP-MS and PGAA is less than 2 ppm. K. Kasviki *et al.* [46] developed a PGAA device that can be used to measure total body nitrogen and protein content in large-volume biological samples or small animals, which can be used to evaluate medium protein in 0.25 - 1.5 Kg biological samples. The Russian MEPhI [24] reactor established a PGAA device for the analysis of element content in BNCT, and the detection capabilities of B and Gd in biological samples reached 1 μg and 10 μg , respectively. Ac-

curate determination of boron content in biological samples is very important in boron neutron capture therapy. The Korea Institute of Atomic Energy [47] used the PGAA system to determine the boron content in mice, and used the standard solution of boron to study the boron content. The solution was applied intraperitoneally to the induced mouse cancer cells and tumors at a dose of 750 mg/kg body weight. The boron content of the two samples was compared with the boron phenylalanine of the tumor and the ethylamine derivative that induces cancer cells, and their accumulation rates in each organ such as blood, spleen, liver, kidney and brain were studied. Analytical quality control by using certified reference materials such as peach, apple and spinach leaves. The relative error of the measured value is in good agreement with the certified value within 2%, and the concentration of boron can be measured to 10 mg/Kg.

4.4. Archaeology

Because PGAA is a non-destructive testing technology, its application in archaeology has become particularly important. Foreign laboratories have repeatedly applied PGAA technology to archaeology.

M. Isabel Prudencio [48] *et al.* analyzed the Lisbon painted tile “The Panoramic View of Lisbon” by combining PGAA, INAA and XRD techniques. The data of thirteen oxygen compounds and elements such as MgO, Cl and B were obtained by PGAA technique. Bogdan Constantinescu *et al.* [49] carried out PGAA and PIXE analysis on ancient glass relics respectively. The “milli-PIXE-PGAA” method is a good supplement to PGAA-PIXE method, which can be used for quantitative and qualitative analysis of SiO₂, alkali flux, stabilizer and colorant in glass components. Ralf Schulze *et al.* [50] used a combination of neutron imaging (NT) and prompt gamma imaging (PGAI) techniques to show the distribution of elements such as Fe, Cu, S, and H in artifacts. K. T. BIR [51] *et al.* analyzed prehistoric stone artifacts. PGAA can distinguish obsidian and felsic porphyry, so as to identify stone artifacts from different sources. However, it is difficult to identify samples with high SiO₂ content and low other elements content, so other methods are needed. The composition of 110 samples, including 76 archaeological works, was reported by compiling data sets from different PGAA analysis series.

4.5. Cosmic Chemistry and Geochemistry

H. R. Marschall *et al.* [52] carried out PGAA analysis of high-pressure metamorphic rocks and compared the data with those obtained by XRF and ICP-MS analysis. For major elements and Gd, Sc, PGAA and XRF, ICP-MS results are in good agreement, for B, Cl, H three elements are very sensitive. Gméling *et al.* [53] analyzed volcanic rocks by combining NAA with PGAA. The experimental results show that the relative deviation of the measurement results of the main elements is less than 10%. For trace elements, most of the measurement results of NAA are accurate, so the two methods can be used to analyze most of the

elements in geological samples. PGAA is also suitable for the analysis of precious samples such as meteorites due to its non-destructive analysis properties and the samples can continue to be used for other methods after PGAA analysis. N. Ahiral [54] *et al.* analyzed meteorite samples by combining PGAA with INAAA, and compared with the results obtained by wet chemical analysis. It is considered that the combination of PGAA and INAA is the most suitable method for the analysis of meteorites. Lea Canella *et al.* [55] analyzed the meteorite samples by PGAA, prompt gamma ray imaging (PGAI) and neutron tomography (NT), and obtained the two-dimensional distribution of Mg, Si and Fe in the samples. At the FRM II research reactor, the first comprehensive analysis of the Allende meteorite was performed using PGAA prompt gamma-ray activation imaging (PGAI) and neutron tomography (NT) techniques. The bulk elemental composition of heterogeneous meteorites was determined by PGAA method. Due to the small size of the sample, only the position-sensitive PGAI analysis was used to obtain the 2D element distribution of the object. As an example, 2D diagrams of Si, Fe and Mg are given. Neutron tomography of meteorites is performed using the same cold neutron beam.

5. Conclusion

PGAA can detect most of the elements on the periodic table of elements, especially for the detection of light elements, which has great advantages. At the same time, it also has the property of non-destructive testing, which can be applied in various fields and has broad development prospects. However, the detection sensitivity is lower than that of neutron activation analysis. By transforming the experimental facilities, such as shielding the background and using the anti-Kang spectrometer, the detection ability of the system can be improved. With the completion of the PGAA facilities of CARR, China will make new progress in this field.

Conflicts of Interest

The authors declare no conflicts of interest.

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