

Overview of Industrial Materials Detection Based on Prompt Gamma Neutron Activation Analysis Technology

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Abstract

Prompt gamma neutron activation analysis (PGNAA) is a non-destructive online measurement nuclear analysis method. With its unique advantages, it has been widely used in online analysis of industrial materials such as coal, cement, and minerals in recent years. However, there are many kinds of literature on PGNAA in the field of industrial materials detection, and there are still a few concluding articles. To this end, based on the principle of PGNAA online analysis, the status quo and development of the real-time online detection of industrial material components in the field are reviewed and discussed by consulting a large number of domestic and foreign PGNAA related literature and data, to facilitate the reference of relevant scientific researchers.

Keywords

Prompt Gamma Neutron Activation Analysis Method, PGNAA, On-Line Detection of Industrial Materials

1. Introduction

Industrial material composition real-time online detection technology is a real-time and accurate online analysis and detection technology for material composition in the process of industrial feeding and batching. It can be widely used in industrial production processes such as coal, cement and minerals. It is an important means to promote the upgrading and transformation of traditional heavy industry enterprises and to energy-saving and emission reduction.

At present, the main means of real-time online detection of industrial material components are: X-ray fluorescence analysis technology, infrared analysis, neutron activation analysis (NAA) and prompt gamma neutron activation analysis

technology (PGNAA). Among them, due to the limitations of the characteristics of the radiation source used, X-ray fluorescence and infrared analysis techniques can only analyze the composition of the surface layer for bulk materials, but cannot perform volume analysis. NAA technology can be used for non-destructive measurement with high sensitivity and good accuracy, but it cannot be used for real-time online analysis. Moreover, it is more powerless for the determination of some light elements (such as H, B, N, P, etc.) [1]. The PGNAA technology has a significant advantage in the sensitivity of non-destructive light element analysis. At the same time, because both neutrons and γ -rays have strong penetrating capabilities, the entire element information of bulk materials can be obtained, so this technology has become the best choice for industrial bulk material composition testing needs [2] [3] [4]. **Table 1** compares the four online analysis technologies.

Prompt Gamma Neutron Activation Analysis (PGNAA) is a non-destructive, efficient, multi-element online nuclear analysis technology [5]. In principle, it can measure most of the elements on the periodic table [6], especially in the analysis of elements with large (n, γ) reaction cross-sections (B, Cd, Gd, H, etc.), its sensitivity is high, and it is usually used as a supplement to conventional analysis, the analysis of light elements in large samples is the only method [7].

Based on relevant international studies, this paper discusses PGNAA technology from the following four aspects: The first part briefly summarizes the basic principle of PGNAA technology and its development direction in recent years; The second part introduces the theoretical research of PGNAA technology in industrial material detection. Further, in the third part of the paper, it mainly discusses the international research on the application of PGNAA technology in industrial material detection system equipment. Finally, in the conclusion part, it summarizes the industrial application of PGNAA technology.

2. The Principle and Development Direction of PGNAA Technology

2.1. Basic Principles

The technical principle of Prompt gamma neutron activation analysis (PGNAA) is shown in **Figure 1**. When the neutron beam emitted by the neutron source bombards the nucleus of the measured substance, undergoes a nuclear reaction (mainly inelastic scattering nuclear radiation capture) and emits instantaneous characteristic gamma rays in a very short time. These characteristic γ are collected through the detection system, and qualitatively and quantitatively analyzing the elemental composition and content of the measured material by analyzing the energy and intensity of these characteristic γ rays [8].

In PGNAA technology, the intensity of γ rays with a certain energy can be given by the following formula [9]:

$$A(t) = \phi \frac{\omega}{M} N_A \alpha \sigma \lambda \varepsilon$$

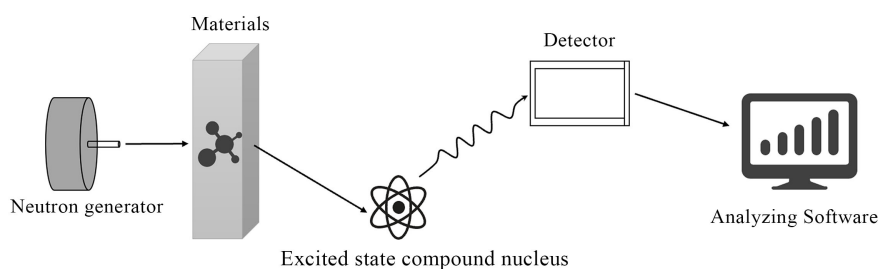


Figure 1. The principle of the Prompt gamma neutron activation analysis technique.

Table 1. Comparison of four online analysis technologies.

Project	Infrared technology	X-ray fluorescence analysis	Neutron Activation Analysis	Prompt Gamma Neutron Activation Analysis
Simultaneous measurement of multiple elements	Yes	Atomic number > 12	Yes (Except H, C, O, etc.)	Yes
Granularity requirements	Have higher requirements	>6 mm	None	None
Influence of surrounding environment	Affected by dust, humidity, etc.	None	None	None
Real-time online analysis	Yes	Yes	Offline measurement	Yes
Measurement method	Surface	Near surface	Volume measurement	Volume measurement
Accuracy	Poor	Poor	Very high	Good
Regular correction	Yes	Yes	No	No
Radioactivity around the instrument	None	Within safe range	Within safe range	Within safe range

$A(t)$ is the count of a certain energy γ -ray in time t ; ϕ represents the neutron flux; ω represents the mass of the detected element; M represents the atomic weight of the detected element; N_A represents the Avogadro constant; α represents the isotopic abundance of the detected element; σ represents the reaction cross section of the element and neutron; λ represents the branching ratio of γ rays; ε represents the detection efficiency of the all-energy peak.

2.2. Development Direction of PGNAA Technology

Since the first use of PGNAA technology to detect elements in 1936, the application research of PGNAA technology has started abroad [10] [11] [12]. In recent years, with the development of nuclear electronics instruments, the detector's ability to detect γ rays has been continuously improved [13], and the rapid development of technologies such as neutron beam devices [14] [15] [16], nuclear databases [6] [17], and energy spectrum analysis [18] [19]. The prompt gamma neutron activation analysis technology has been widely used in the analysis of

multi-element composition of large samples in many disciplines including environmental science [20] [21] [22], industry [23] [24] and health sciences [25] [26]. At this time, with the development of radioisotope neutron sources [27] and portable neutron generators [28], the technology has been able to conduct in-situ and rapid online analysis of large samples such as mining [29] [30], biological [31], manufacturing and construction quality control tasks [32] [33] [34], as well as homeland security contraband detection [35].

In recent years, with the widespread application and in-depth development of PGNAA technology, the international research hotspots mainly include the following three aspects:

1) PGNAA system methodology [36]: In the field of methodology, the research of prompt γ neutron activation mainly includes the study of k0 method in the quantitative analysis of PGNAA technology [37] [38], the methodological study of the calibration of the wide energy range γ -ray detection efficiency [39], as well as evaluation and analysis techniques of γ energy spectrum [19] [20] [40], etc.

2) PGNAA system device design: Since the late 20th century, with the progress and development of electronic technology and detection systems, the current development trend of PGNAA technology has also changed from increasing the neutron fluence rate to increasing the lower detection limit of elements, etc. On the other hand, compared with hot neutron beams [41] [42], the cold neutrons have a higher reaction cross-sections and lower energy; at the same time, due to the small fraction of fast neutrons in the cold neutron beam and the low γ -ray interference [43], the design and research of the cold neutron beam system with high fluence rate has become a hot spot in the world [43]-[48]. For example, in the US National Institute of Standards and Technology [49] [50] [51], Japan Atomic Energy Research Institute [52], Korea HANARO [53] [54] and other reactors have established their own advanced PGNAA system devices. In China, there are currently two PGNAA facilities in operation. One is prompt gamma neutron activation analysis (PGNAA) system based on the In-Hospital Neutron Irradiator (IHNI) developed for the Boron neutron capture therapy (BNCT) project on a 30kW micro research reactor in Beijing [55]; the other is based on the Prompt gamma Neutron Activation Analysis (PGNAA) system on the China Advanced Research Reactor (CARR) [56] [57]. In recent years, in order to be able to irradiate the target sample with a higher neutron fluence rate to improve the sensitivity of PGNAA system analysis, a focused neutron beam analysis method has been proposed internationally [58] [59]. At the same time, the detection technology of Compton suppression gamma spectrometer was developed to improve the detection defects of low-energy gamma rays, which greatly reduced the Compton effect and improved the detection efficiency of low-energy gamma rays [60] [61].

3) Application of PGNAA technology: As a high-sensitivity, multi-element non-destructive online analysis method, PGNAA has been widely used in more

than 30 PGNAA facilities worldwide. In recent years, PGNAA technology has proved to be valuable in a variety of applications, and its research field covers many aspects such as materials science, biology and medicine, earth science and cultural research.

3. Application of PGNAA Technology in Industrial Material Inspection

The use of the prompt gamma neutron activation analysis (PGNAA) method for the on-line analysis of the total element composition of industrial materials has been a hot spot in the field of nuclear detection and analysis in recent years [62]. Its application is mainly aimed at resource-intensive and high energy consumption industries such as coal, cement and metallurgy. This technology is used to conduct real-time detection of material composition in the production process so as to timely adjust the ratio of materials, so as to realize the optimization of the production process and ultimately improve the production efficiency [63]. For example, the coal blending process of thermal power, and the raw material proportioning process of cement and metallurgical industries can all use online testing equipment to reduce fuel consumption, improve production efficiency, and control product quality.

3.1. Online Analysis and Detection of Coal

Online detection of coal has been studied in the United States since the late 1970s. [1]. At the same time, its research and development equipment was applied to the Homer City Coal Preparation Plant in the United States in the 1980s to detect the low sulfur and low ash products of the coal preparation plant. Then developed Nucoalyzer, a coal detection device for power plants, which is based on ^{252}Cf neutron source and a set of high count rate spectrum processing system for online detection and analysis of coal [64]. Because the average energy of the neutrons emitted by the ^{252}Cf neutron source is low, it mainly reacts with the elements in the material to capture the thermal neutron, and the detection of key elements such as C and O in coal cannot be achieved [65]. Therefore, in 1995, Wombel *et al.* proposed a method for online analysis and research of coal using pulsed fast-thermal neutrons [66]. Subsequently, L. Dep *et al.* made further research on this basis, using pulsed neutron generator and BGO detector to realize the online analysis of major and minor elements in coal. The results showed that the accuracy of carbon in the sample can be nearly 1%, and the accuracy of sulfur reached 0.05% [67]. In 2001, Australia's M. Borsaru *et al.* applied PGNAA technology to coal borehole logging and measured the density of fly ash, iron, silicon, aluminum and coal seam downhole [68]. Later, the ^{252}Cf neutron source and BGO detector were used to study the sulfur content in the coal seam [69]. From the 1990s, Thermo Electron Corporation of the United States carried out research and development of Coal analyser. Combined with PGNAA and PFTNA (Pulse Fast-Thermal Neutron Analysis) technology, it de-

veloped the second-generation Coal Quality Manager FLEX, which can conduct accurate qualitative and quantitative analysis of S, Si, Al, Fe, Na and other elements and water content in Coal.

3.2. Total Element Analysis of Cement Materials

In the cement industry, Argentine scholar Daniel L. and others pioneered the analysis of elements in cement samples, and the analysis results showed that PGNAA technology could achieve the measurement of elements such as Fe, Ca, Si and Cl in the samples [70]. In 1999, R. Kheli *et al.* used Am-Be neutron source and high-purity germanium detector to measure the ratio of silicon to calcium in cement samples [71]. Saleh H. *et al.* have developed a device for detecting the content of chlorine in reinforced concrete [30]. In 2001, C.S. Lim *et al.* have developed the PGNAA cement on-line testing equipment on the conveyor belt. The equipment utilizes the inelastic scattering and capture reaction of the neutrons emitted by the Am-Be neutron source and the elements in the sample to realize the analysis of cement raw material elements, and the dual source detector is adopted to reduce the measurement error caused by uneven spatial distribution of the raw material components on the belt [72]. From 2009 to 2014, A.A. Naqvi *et al.* successively studied the cement dust and the elemental chlorine in cement, and analyzed the cement dust and concrete by the PGNAA technology to obtain the detection limit of elemental chlorine [73] [74] [75].

3.3. Applications in the Mineral and Metallurgical Industries

The iron and steel industry is an important pillar industry for national construction and national economic development, and is a basic industry for industrialization, as well as an intensive consumption of technology, capital, resources, and energy. The traditional detection techniques are mostly X-ray fluorescence analysis, neutron activation analysis (NAA), etc., and with the help of the high-sensitivity analysis performance of PGNAA technology, research teams in various countries have begun to use PGNAA technology to carry out elements in various ores. Qualitative and quantitative analysis, as well as quality inspection in the production process of iron and steel smelting. During the metal smelting process, the presence of hydrogen may cause the brittleness of the material to increase, which has a great impact on the performance of the steel, so it has always been the focus of attention [76]. The Cold neutron instantaneous gamma activation analyzer at the National Institute of Standards and Technology (NIST) Center for Neutron Research (NCNR) has been shown to detect trace hydrogen in advanced materials such as titanium alloys [77]. In 2010, H.J. Cho *et al.* used PGNAA technology to analyze the hydrogen element in low-alloy steel and obtained good test results [78]. In 2017, the Danyal team used PGNAA technology to evaluate the performance of trace hydrogen elements in titanium alloys, and the uncertainty range was consistent with the experimental values [79]. In the application of ore element analysis, in 2009, Nicola's team used the PGNAA method to detect and analyze the Cl element in the ore sample. The re-

sults showed that the uncertainty of the Cl element content was less than 5%, which was the element of the ore. The analysis provides new technical methods [80].

3.4. Domestic Research on PGNAA Material Detection Technology

The on-line detection and analysis of the material composition of PGNAA technology first started in Europe and America and other developed countries. In recent years, with the vigorous development of nuclear technology applications, the domestic market has slowly started. In 1996, Chen Boxian *et al.* conducted a preliminary analysis of coal using the Am-Be neutron source, which provided a feasibility study for PGNAA technology to analyze the main elements in coal [81]. Subsequently, Song Zhaolong, Jia Wenbao and other teams have successively carried out the research of PGNAA technology in the field of online coal detection, and achieved remarkable results [82] [83]. In terms of elemental analysis of cement, Cao Xuepeng's team used D-D neutron generator as the neutron excitation source to carry out relevant research on the main elements of cement raw materials. The relative deviation of the measurement results is less than 7%, which can meet the requirements of industrial production [84]. For the application of ore detection, Song qingfeng's team used PGNAA online analysis technology to measure the copper and nickel elements in the ore, and the detection accuracy can meet the requirements of online analysis of iron ore in the industrial site [85]. Then, through the combination of simulation and experiment, the team tested and verified the composition of iron ore pulp, and the results showed that the fitting degree of the experimental determination and analytical value of each component exceeded 0.99, proving the feasibility of PGNAA technology in the analysis of pulp composition [86].

4. Development and Application of Industrial Online Detection System Based on PGNAA Technology

The real-time online detection system of industrial material composition is mainly based on the PGNAA technology of isotope neutron source or neutron generator. By detecting the prompt γ -rays generated by the nuclear reaction between neutrons and nuclide in the material sample, and by online data analysis to obtain the element composition and content of the test object. A complete PGNAA online analysis system mainly includes three parts: field test, signal processing, data transmission and feedback. Among them, the on-site testing mainly includes detectors, neutron sources (or neutron generators), measuring mechanisms and protective devices; the signal processing part includes the spectrometer system, spectrometer analysis and data processing system; the data transmission and feedback part mainly includes the results display and data exchange of analyzer, industrial control unit, etc. [3].

4.1. Overview of Foreign Industrial Online Detection Systems

The traditional PGNAA industrial material online analysis system is mainly de-

veloped based on isotope neutron source. In recent years, with the advancement of micro neutron generators and its unique advantages, it has gradually replaced the isotope neutron source and is used in the PGNAA industrial material online analysis system [1]. In the late 1980s, the original gamma metrics company (now Thermo Fisher Scientific) of the United States developed the first elemental composition analyzer for industrial sites after decades of technical research, and successfully applied it to the coal and cement industry [87]. At present, there are three major companies in the world that manufacture PGNAA systems for on-line real-time analysis of industrial material components: Thermo in the United States, Scantech in Australia, and Sodern in France [88]. The CB-Omni, which was launched by Thermo of the United States in 2006, is based on the isotope ^{252}Cf neutron source. The detection system uses two NaI detectors, and its application field is mainly oriented to the mining and cement industry [87]. Scantech in Australia has mainly developed the COALSCAN 9500 analyzer for the coal industry and the GEOSCANTM analyzer for the cement industry. Both analyzers use ^{252}Cf as the neutron source, among which, COALSCAN 9500 uses NaI detector as the detection system, which can perform complete online analysis of coal [89]; and GEOSCANTM uses multiple BGO detectors as the gamma rays of the device, and the detection system has a high γ photon counting rate. The CAN device of Sodern in France uses an electrically controllable D-T neutron generator as the neutron excitation source. Since the D-T neutron generator can provide neutrons with higher energy, it can generate inelastic scattering reaction with C, O and other elements captured in the material with smaller cross sections to stimulate the prompt γ rays. Therefore, the device can realize full-element analysis, and its analysis index is more comprehensive. At the same time, based on the advantage of neutron source electrical controllability, it can provide a safer environment for equipment maintenance [90].

4.2. Research and Application of Domestic Industrial Material Detection System

Now in Europe and America and other developed countries, the development and manufacturing of PGNAA technology-based real-time online detection system for industrial material composition has already formed a relatively mature industrial chain, and is sold through relevant enterprises. With the development of our country's industrial technology, domestic demand for real-time online detection systems for industrial materials has gradually increased. In the early 1990s, aluminum plants such as Shandong and Zhengzhou began to use the NH-501 system based on PGNAA technology for industrial online test and measurement [3]. Since the beginning of this century, the domestic market has become active. Based on the use of foreign equipment, Nanjing Continental Technology has achieved localization through innovation and reconstruction. Afterwards, scientific research units such as Kelun Company of Northeast Normal University, China Academy of Engineering Physics, Tsinghua University and China Atomic Energy Research Institute also launched related PGNAA

projects [24]. Based on the modern nuclear analysis technology, a series of testing equipment for the analysis of industrial components such as coal and cement has been developed and produced. Its quality and technology have made greater progress than before, but compared with similar products abroad, there is still a big gap in performance. High-sensitivity, multi-parameter measurement of high-end industrial material composition real-time online in-situ detection system still needs to rely on foreign imports.

Based on the good application prospects of PGNAA technology in China's industrial material detection technology, the National Ministry of Science and Technology established a special project for the development of national major scientific instruments and equipment in 2013: "Real-time online detection of industrial material components based on prompt gamma-ray neutron activation analysis technology System". The project is led by Nanjing University of Aeronautics and Astronautics, where Academician Chen Da's team is located, and cooperates with 8 units including Lanzhou University and Northeast Normal University. It aims at energy conservation and consumption reduction in China's resource-intensive and consumable industries, from national industrial development and material composition. Starting from the new trend of analysis technology, developed a real-time online detection instrument for industrial material components with national independent intellectual property rights and reaching the international advanced level. The instrument is based on the prompt γ neutron activation analysis technology, uses the neutrons generated by the controllable small D-D/mini D-T neutron source, after being modulated by the slowing multiplication system, bombards the measured material to produce prompt characteristic γ rays, using the new high-efficiency γ -ray detection system obtains the γ energy spectrum and analyzes it to obtain the main elements and their contents in the material [91]. In this project, the two key technologies at the core are: the development of high-yield, long-life D-D small neutron sources, miniaturized of a long-life D-T sealed neutron sources, and the development of high-count rate detection systems [92]. At present, the project has completed the application development research in the fields of thermal power plants, ironworks, cement plants, etc., has built three demonstration projects, and passed the self-acceptance phase in December 2018.

5. Summary

As an effective nuclear analysis technique, prompt gamma neutron activation analysis plays an important role in the on-line analysis of industrial materials, especially in the detection of irregular bulk materials. Nowadays, PGNAA technology is relatively mature in terms of principles and basic applications. In recent years, international scientific research teams have carried out further research in the development of new detectors such as high resolution, development of energy spectrum analysis and application development of portable neutron sources. On the other hand, the neutrons and gamma rays arising from the

neutron source are severely harmful to the human body. Processing and optimizing the various parameters of the PGNAA system setup under the irradiation of a neutron source is dangerous and time consuming. The Monte Carlo method has a huge advantage in complex particle transportation. Therefore, many scholars have studied the Monte Carlo method, which has become a hot spot in the world.

In summary, after more than half a century of development, the application of the instantaneous gamma neutron activation analysis technology in the detection of industrial materials has reached a relatively high level in developed countries such as Europe and America. With the strong support of the country and the unremitting efforts of the scientific research team, the technical level of PGNAA in China has made remarkable progress, but there are still gaps. It is necessary to continuously realize the improvement of technology, strengthen the in-depth integration of production, education and research, and localize the PGNAA technology. And maturely applied to the online detection of industrial materials.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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