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중성자 핵반응을 이용한 원소 검출기술
- 즉발감마선 중성자 방사화분석법을 이용한 검출기술 -

송병철★ · 박용준 · 지광용
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Elemental Analysis by Neutron Induced Nuclear Reaction
- Prompt Gamma Neutron Activation Analysis for Chemical Measurement -

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요 약 : 즉발감마선 중성자방사화법 (PGAA)은 시료내 미량 및 주원소를 빠르게 비파괴적으로 분석하는 방법으로 주로 광물, 금속, 석탄, 시멘트, 석유, 코팅, 제지 등 다양한 산업체에서 실시간 분석법으로 매우 유용하다. 이 방법은 제약과 관련된 산업체 또는 연구업무에도 활용되며, 마약 또는 폭발물과 같은 위험물질의 탐지에도 이용되고 있다. 본 총설은 즉발감마선 중성자 방사화법의 현재의 기술현황과 앞으로 연구추진 경향에 대하여 서술하였다. PGAA 시스템은 중성자 선원, 중성자 핵반응으로부터 발생하는 즉발감마선을 측정하기위한 다중채널분석기와 A/D 변환기 등의 전자모듈과 고분해능 HPGe 검출기로 구성된다. 속중성자의 콤프턴 산란에 의한 높은 바탕값은 감마-감마 동시계수장치의 도입으로 개선될 수 있다. 현재 ^{252}Cf를 사용한 즉발감마선 중성자 방사화 장치는 수용액중에 존재하는 원소들의 실시간분석을 위해 한국원자력연구소에서 개발중에 있다. 이 장치는 다양한 마약 및 폭발물 또는 화학무기의 탐지에도 응용될 수 있다.

Abstract : Neutron induced prompt gamma activation analysis (PGAA) offers a nondestructive, sensitive and relatively rapid method for the determination of trace and major elements and is proven to be convenient for online analysis of minerals, metals, coal, cement, petrochemical, coating, paper as well as many other materials and products. The technique has found many uses in medicine, industry, research, security and the detection of contraband items. This report reviews the present status and future trends of the PGAA techniques. Requirements for the system are neutron source, high resolution HPGe detectors with a high-voltage power supply, an amplifier, analog-to-digital converter, and a multichannel analyzer for the detection and measurement of prompt γ-ray emit form the neutron capture elements. Introducing a γ-γ coincidence system also improves the quality of the γ-ray

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spectrum by suppressing the background created from the Compton scattering of high energy prompt γ-rays. A PGAA system using a ^{252}Cf neutron source is currently under construction for the on-line measurement of several elements in aqueous samples at KAERI. The system can be applied for the detection of chemical weapons and explosives as well as various narcotics.

Key words : prompt gamma, thermal neutron, γ-γ coincidence, elemental analysis

1. Introduction

TNeutron induced prompt gamma activation analysis (PGAA) is a recently developed nuclear analytical technique for qualitative and quantitative analysis of elements. Prompt gamma ray is generated by the interaction of neutrons from research reactors or isotopic sources with a target element. The technique is suitable for in-situ measurements and bulk sample analysis. PGAA is applicable for light elements and for certain radionuclides. This technique is a complementary analytical tool for delayed neutron activation analysis and can provide substantially higher sensitivities than the instrumental neutron activation analysis (INAA). Analytical techniques based on nuclear interaction are well established and widely used such as INAA and radiochemical neutron activation analysis (RNAA), or in a vigorous development stage such as PGAA. Since these nuclear analytical techniques are non-destructive, sensitive and also particularly quick, they are used for on-line analysis of materials. The industries in which nuclear techniques for on-line analysis are used are mineral processing, metallurgical, industrial minerals, coal, petrochemical, coating and the paper industries.

It is not surprising that the penetrating neutron can carry information of elemental composition from deep parts of the sample to be measured by passing through the matter being analyzed and inducing prompt gamma-ray through either the thermal neutron capture or fast inelastic scattering processes. As a result, thousands of batch samples can be analyzed directly on the conveyer in a short time period without tedious preparation prior to analysis. Applications of prompt gamma-ray technique to various industries for on-line bulk analysis of coal, mineral ore, nitrogen-contained explosives, as well as geothermal water have proven to be an attractive alternative to the conventional methods.[1]

This report reviews the basics of neutron induced nuclear analytical techniques, development of neutron induced prompt gamma activation analysis and the present status of PGAA.

2. Basis and development of PGAA

At present there are two methods for the analysis of elemental composition using neutron-induced reactions, they are NAA and PGAA. These nuclear analytical techniques are based on the interaction of neutrons with matter. As a consequence of this reaction, there is usually the formation of two products; the light one and a heavier product. The light one is normally γ-ray or neutrons. The interaction of neutrons with matter is practically confined to the nuclei and comprises of elastic and inelastic scattering and nuclear reactions. In elastic collisions the total kinetic energy remains constant, whereas in inelastic collisions parts of the kinetic energy are given off as excitation energy.

In order to understand neutron interaction, it is convenient to introduce the scattering cross section. For a very thin target, the number of scattered particles is very small compared with the number of particles in the beam and

$$-\frac{dN}{N} = \frac{(nA\,dx)}{N} = n\sigma dx \quad \text{---------- (1)}$$

where dN is the number of scattered particles, N is the number of incoming particles, dx is the thickness of

the target, and A is proportionality constant dependent on the number of nuclei per unit volume n and the nature of the nuclei represented by σ, the scattering cross section, which is a measure of the probability that a nuclear interaction between the incoming neutron and the nucleus of a particular atom will occur. The unit for the cross section is the barn and is equal to 10^{-24} cm^2. *Table* 1 shows the captured cross sections and the prompt gamma-rays of various elements that can be typically measured by PGAA.

Table 1. Characteristics of selected capture gamma-rays for various elements

Element	Nuclide	Isotope ratio (%)	Capture Cross section				Prompt γ-ray	
			thermal (mb)		fast (mb)			
			nuclide	average	nuclide	average	energy(keV)	intensity
B	B-10	19.9	500.0		0.021		4443.0	76.0
	B-11	80.1	5.075		---		6759.3	39.5
							7005.1	47.9
Mg	Mg-24	79.0	50.29		0.034		585.2	52.6
	Mg-25	10.0	190.4	62.98	0.007	0.030	1808.9	61.6
	Mg-26	11.0	38.31		0.021		2828.1	87.4
							3916.7	100
Al	Al-27	100	231.1		0.253		248.9	6.8
							2960.0	8.0
							7723.9	27.4
Si	Si-28	92.2	176.7		0.706		1273.3	15.9
	Si-29	4.67	101.4	171.1	0.325	0.681	2092.9	21.5
	Si-30	3.10	107.5		0.482		3539.1	68.0
							4934.4	62.7
Ca	Ca-40	96.9	407.5		0.500			
	Ca-42	0.65	683.0		0.001		1942.0	72.6
	Ca-43	0.14	11660	435.8	0.0002	0.485	4418.9	14.9
	Ca-44	2.09	888.4		0.001		6419.9	38.9
	Ca-46	0.004	740.0		0.001			
	Ca-48	0.187	1093		0.002			
F	F-19	100	9.570		0.026		582.2	13.0
							2452.8	10.2
							3589.3	11.1
Cl	Cl-35	75.77	43640	33140	0.349	0.334	516.7	18.5
	Cl-37	24.23					788.4	15.0
							1164.7	19.9
							1950.9	21.7
							1959.1	14.6
							6110.9	20.0
							7413.8	10.4
S	S-32	95.0	528.2		0.003		841.1	75.6
	S-33	0.75	350.0	513.9	0.001	0.003	2379.7	44.5
	S-34	4.21	223.6		0.004		3220.8	27.1
	S-36	0.02	150.0		0.001		5420.5	59.1

Neutrons are generally classified as their kinetic energies. There is no sharp division or energy line of demarcation between the various classes of neutrons; however, the following is an approximate categorization according to its energy: cold; < 0.003 eV, thermal; 0.003 - 0.4 eV, epithermal; 0.4 - 100 eV, intermediate; 100 eV - 200 keV, fast; 200 keV - 10 MeV and high energy neutrons; > 10 MeV. Neutrons with energies in the range of 1 eV to 1 keV are also called resonance neutrons, because the maxima of absorption is observed in this energy range.

A fast neutron which loses its energy mainly in elastic and inelastic collision suffers many interactions in the process of losing its energy to reach thermal energy. This process is called thermalization. The neutron may disappear during this process due to nuclear reactions when the neutron impinges on nuclei forming the scattering medium. The probability of scattering from or reaction with a single nucleus is called the cross-section, as mentioned previously. Collision of neutrons with nuclei can result in scattering of the neutrons and recoil nuclei with a conservation of momentum (elastic scattering) or loss of kinetic energy for the neutron as gamma radiation (inelastic scattering). The capture of neutrons by a nucleus of an atom may result in the emission of other nuclear particles from the nucleus (nonelastic reactions) or the fragmentation of the nucleus into two (nuclear fission).

After the development of a neutron guide tube and a cold neutron source in late 1960, the first PGAA experiment using a thermal neutron guided beam was carried out at the Saclay Center for Nuclear Studies in France in 1968. In 1973, a highly sensitive PGAA experiment for multi-element determination and its application to geological samples was carried out by Henkelmann and Born using the high flux cold neutron guided beam of the HFR reactor at the Institute of LaueLangevin (ILL) in France. In 1983, Kobayashi and Kanda constructed a compact PGAA system using the thermal neutron guided beam and LiF tiles as neutron shielding material at the Kyoto University Reactor in Japan for the determination of ^{10}B in boron neutron capture therapy (BNCT). In 1987, two experimental results on the development of a PGAA system for the FRJ-2 reactor of the Julich Research Center in Germany and HFR reactor of the ILL in France were published.[2] After that, the PGAA system for analytical purposes with cold and thermal neutron guided beams became widely available in the 1990s. Now PGAA research is widely conducted using cold or thermal neutron beams by several institutes, such as JAERI in Japan, NIST and the University of Texas in the USA, the institute of isotopes (IKI) in Hungary, which is the only institute with a research reactor for PGAA in Europe.[2]

The concept of a portable PGAA system consisting with a ^{252}Cf neutron source is very recent and a rapidly developing analytical method. A number of organizations developed the system for on-line characterization of cement and other bulk raw materials,[3] analysis of bulk coal,[4] borehole logging for in-situ elemental analysis for the coal and mineral industries.[5] A portable PGAA system consists of ^{252}Cf neutron source subsystems, a HPGe detector and a portable MCA integrated with a laptop computer and was developed at the Turner-Fairbank Highway Research Center, McLean, VA, USA to analyze the elemental composition of reinforced concrete and to measure chloride contamination.[6]

3. Techniques and instrumentation in PGAA

3.1. Neutron source and its assembly

When a material is bombarded with neutrons, high energy γ-rays of different energies are produced. A detailed description of the PGAA technique is available in recent books edited by Alfassi and Chung.[1] PGAA involves bombarding the material to be analyzed with neutrons and measuring the characteristic γ-rays produced by the elements in the sample. After the nucleus is in an excited state by neutron capture, one or more prompt γ-rays with specific energy is released within 10^{-14} seconds. The emission rate of this γ-ray from a sample in a neutron beam can be used to

Table 2. Thermal neutron attenuation factors for various neutron shielding materials

Shielding Material	Macroscopic Thermal Neutron Cross-Section	Thickness		Attenuation Factor (I_0/I)
		inches	mm	
Water	0.02	1	25	1.05
Pure Polyethylene	0.03	1	25	1.07
5% Boron-Polyethylene	2.00	1	25	161
80% Pb, 1% Boron-Polyethylene	1.80	1	25	97
10% Boron-Putty	4.70	1	25	105
Boron Flexible Sheet (25.3 %)	17.50	0.125	3	259
30% Boron-Polyethylene	14.50	1	25	1016
7.5% Lithium-Polyethylene	0.48	1	25	3.4
Boro-Silicon (1%, castable)	0.71	1	25	22

measure the amount of element in the sample. A mixture of elements gives a mixture of many γ-rays, so the capture spectra are complex, often containing several hundred peaks. The neutron collimator and neutron shield can be made from a combination of both thermalization and absorption materials. Ideal thermalization materials should contain light elements with large scattering and small absorption cross-sections for fast neutrons. Water, heavy water, beryllium, graphite, paraffin and various polymers are the most effective media to reduce the energy of the neutrons. About 20 collisions with protons are necessary to slow a neutron of several MeV down to thermal energies. For this purpose, about 20 cm of paraffin is necessary. Graphite is also used as a moderating material for neutrons. About 120 collisions with the neuclei of carbon atoms are necessary to have the same effect as 20 collisions with protons. After having lost the main part of their energy, the neutrons are captured by nuclei to give rise to nuclear reactions. Thermal neutron absorption materials are those containing elements with high capture cross-sections, low emission rates of prompt gamma rays, and negligible amounts of induced radioactivity. Among the best choices are materials containing ^{6}Li, ^{10}B, ^{113}Cd and some rare earth elements such as Sm, Eu, Gd, or Dy.

Neutron-flux attenuation is an important function of the shield to protect an analyzer. Thermal neutron attenuation factors for various neutron shielding materials are listed in *Table* 2. In all region of high neutron flux, borated polyethylene is used for the shield in most cases. In critical areas near the detector ^{6}Li compounds instead of borated compounds are often used, since they have a large cross section for the (n, α) reaction, which does not produce γ-ray while the neutron captured by the boron will result in a 0.42 MeV gamma ray and will therefore not completely eliminated the gamma background. The simplest of all is the ^{1}H (n,γ)^{2}H, capture of neutrons by a proton. In this case, the capture state at 2.2 MeV can be decayed by only one way, by emitting a characteristic 2223.23 keV γ-ray.

Most of the ongoing PGAA systems use neutron beams from research reactors, in which beam ports are available to extract neutrons from a high-flux region near the core to an accessible location outside the shielding. PGAA requires thermal neutrons. The ^{252}Cf spontaneous fission source is well suited for the portable PGAA technique. The ^{252}Cf neutron sources of 50 - 200 μg (1.1 - 4.6 $\times 10^{8}$ n/s) are normally used for this purpose. It is possible to use a fast neutron source for PGAA, by slowing down fast neutrons after leaving the source either by an external moderator or by moderation of the sample itself. When analyzing

aqueous samples or coal, the moderation of the sample itself can be achieved especially by using the transmission geometry, where the sample is positioned between the neutron source and the γ-ray detector.

3.2. Detectors and electronics for prompt gamma ray

In order to analyze the prompt γ-ray in PGAA experiments, photon detectors are required. The best common γ-ray detector used in PGAA is the Ge γ-ray detector. The gamma photon reacts with the detector and to produce electrons, which induce excitation or ionization of the Ge crystal. Solid-state detectors are used for its high resolution and a long counting time is required to measure minor elements such as Na and Mg. However, it suffers radiation damage from the neutron interactions in the semiconductor crystal, which ultimately causes permanent degradation in the resolution. In order to reduce the noise due to the leakage current of the electrons in the conduction band agitated by thermal excitation, the semiconductor crystal is cooled to the liquid nitrogen temperature. Of all semiconductor materials, germanium is exclusively used for modern gamma spectrometry since only for Ge and Si can it be prepared in highly pure form. In the mid 1970's, advances in germanium purification technology made available high-purity germanium (HPGe) that could be used for γ-ray spectrometry detection without lithium drifting. HPGe detector must be cooled to 77K during the measurement.

An array of two γ-ray detectors operated as a pair of spectrometer is also common in PGAA applications, since prompt gamma experiments typically have a high level of background radiation. Pair spectrometry with the combination of more than one detector is connected either in an anti-coincidence or coincidence mode for the purposes of either suppressing the Compton scattering or removing ambiguities to identify the original energy of the photon.

The most widely used detection instrument for PGAA is a Compton suppressed spectrometer, which greatly reduces the size of the continuum and of the escape peaks.[7] The Compton scattered gamma rays escaping from a Ge detector can be registered again on another detector whose signal can be used to oppose the signal from the Ge detector, thus yielding a spectrum in which the Compton background as well as single- or double-escape peaks are suppressed. This system is called escape-suppressed spectrometer. In this arrangement the overall sensitivity of the surrounding detector should be as high as possible. A suppression of three to eight is usually obtained. The system is especially useful for the detection of weak low energy transitions in the presence of high-energy gamma rays.

The double-escape peak is due to pair-production followed by two annihilation quanta which have escaped completely from the crystal. Therefore, the detection of the 511 keV-511 keV pair by means of a couple of detectors sandwiching the detector crystal can be used to gate the detector signal in order to eliminate all but the double-escape peaks. This system is called the pair spectrometer. In this coincidence arrangement the absolute sensitivity of the double-escape peaks is easily reduced by the detection factor of the 511 keV-511 keV pair, but the suppression of the other events is almost perfect.

These two alternative methods can be applied simultaneously, to use the surrounding detectors jointly. Alexander et al. used a split annulus of a Na(Tl) crystal surrounding a central Ge(Li) crystal for an accelerator experiment.[8] The efficiency in the pair spectrometer mode was 22%. The improvement in signal to noise ratio was by a factor of 20. Such simultaneous measurements of both pair and escape-suppressed spectra help greatly in the unambiguous identification of complex gamma ray peaks, and also in the relative energy calibration based on the energy difference of 1022 keV for any pair of full-energy and double-escape peaks.

Coincidence detection of gamma rays is a well-established technique in nuclear structure studies.[9,10] In the γ-γ coincidence method there are several important considerations such as relative efficiency of each of the detectors for two γ-rays, possible spurious

coincidences due to the Compton scattering of γ-ray from one detector into the other, possible angular correlation of the γ-ray, and attenuation of this correlation due to the finite sizes of the source and detector. Peelle and Maienschein described the treatment of the problems for low detection efficiencies.[11] Conventional method for undertaking γ-γ coincidence is to require a coincidence relation with a selected full-energy peak. This constraint reduces the spectrum to the signals of those gamma photons which are in a cascade relation with that peak, thus it lowers the background substantially, increasing the peak to background ratio. Gardener et al. recently demonstrated the possibility to eliminate the hydrogen prompt γ-ray background while increasing the signal to noise ratio in PGAA in a qualitative way.[12] A significant disadvantage is that this method is extremely time consuming due to the low coincidence count rate.

Instead of using the peak-coincidence method, Ember et al. proposed a γ-γ regional coincidence method to define a coincidence relation not with a single peak, but with a selected part of the spectrum containing several peaks and a part of their Compton continuum to increase the coincidence efficiency.[13] The spectra of ungated, Compton suppressed and coincidence are compared in *Fig.* 1. The Compton suppression reduces the hydrogen single-escape peak to full-energy peak ratio by a factor of 10, and the continuous Compton background is lowered by a factor of 10 at the γ-ray energy between 1000 and 2000 keV. However, the boron peak and its Compton background still dominate the low-energy part of the spectrum as shown in *Fig.* 2, in which the low-energy part under 550 keV of the same spectra is enlarged. The coincidence method completely removes the hydrogen peak, its Compton edge, and both of its escape peaks. As can be seen clearly in *Fig* 2, the boron peak and its Compton continuum are also reduced substantially, making the low-energy γ-ray much more visible. The large number of peaks over a large energy range require a pulse-height analyzer with at least a 16,000 channel conversion gain. To reduce background from γ-rays

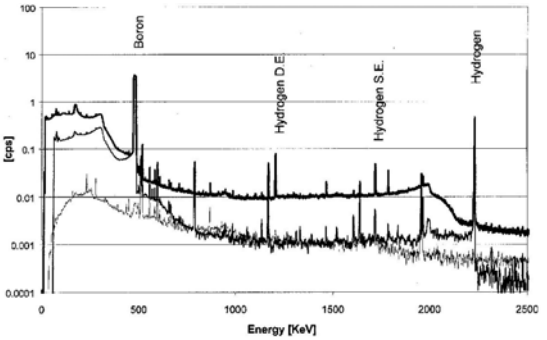

Fig. 1. Comparison of spectra of ungated, Compton suppressed and coincidence. (full energy range between 0 and 2500 keV).

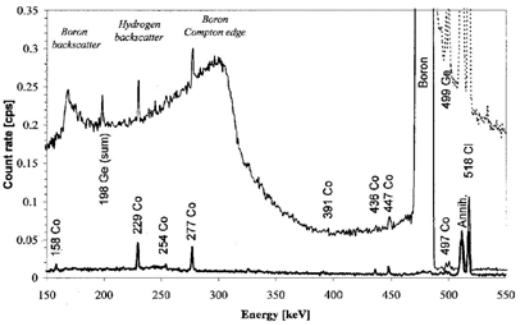

Fig. 2. Comparison of spectra of ungated, Compton suppressed and coincidence. The low energy range under 550 keV of the spectra in *Fig*. 1 are enlarged.

and stray neutrons, the detector must be shielded by lead inside a neutron absorber. The shielding also incorporates thermalizing material to slow the epithermal absorbers. Li enriched in ^{6}Li is often used.

4. Development of database for PGAA

Comprehensive prompt neutron capture γ-ray data is important for PGAA study. PGAA is applicable to all elements including the light elements, usually difficult or even impossible to measure in conventional neutron activation analysis (NAA) due to the lack of suitable radioactive products. In spite of the many advantages of PGAA against NAA such as highly sensitive to certain elements, fast and instantaneous, it suffers from some

disadvantages. The major obstacle for use of the PGAA method has been the lack of a suitable database of characteristic prompt gamma ray energies and intensities so far. Due to the large number of peaks, typical for a prompt gamma ray spectrum, a suitable analytical library must contain very precise energy and absolute intensity data of all the stable elements and their isotopes for performing element identification and quantitative analysis.

The most-used prompt capture gamma ray database for elements is the twenty-year-old compilation by Lone et al.[14] However, this database contains many contaminant lines, misses important transitions, and lacks the precision sufficient for chemical analysis, and therefore does not meet present-day standards.[15,16] The International Nuclear Data Committee of the IAEA, at its 1997 meeting in Vienna, strongly recommended that the Nuclear Data Section supports the update and new measurements of data needed in PGAA.[17] An advisory Group Meeting on the Coordination of the Nuclear Structure and Decay Data Evaluators Network held in Budapest, from 14-18 October 1996 decided to replace the old compilation with a fundamentally new compilation.[18,19] Molnar et al. are to establish a new database that contains all observable prompt gamma lines, as well as the ones from other reactions, with accurate energy and intensity data appropriate for elements identification.[15,20] They have shown that the k_0 standardization method in NAA may be applicable to PGAA. The k_0-factors on the basis of stoichiometric measurements may be used for the determination of relative elemental concentration without the need for standards.[21] An IAEA coordinated research project was formed to combine the Budapest reactor data with isotopic measurements from the literature to create a comprehensive database of thermal capture γ-ray cross-sections and k_0 values. The β-version of this database has now been completed providing PGAA data for 31,000 prompt γ-rays for all the elements from hydrogen to uranium.[22]

5. Applications of PGAA

PGAA is a technique for measuring the elemental composition of bulk materials and well suited for the rapid on-line analysis of major elements in different matrices. The method is most useful for measuring elements that capture neutrons efficiently. PGAA is a complementary method for neutron activation analysis of the following important elements: H, B, C, N, P, S, Cd, Sm, and Gd. Further, PGAA seems to be the best technique for the determination of H, B, Si, S, P, Cl, Ca, Ti, Cr, Mn, Fe and Ni for borehole logging and field analysis.[16] The most commonly used techniques for on-line analysis are divided into photon methods and neutron methods. Photon methods are X-ray techniques based on fluorescence, scattering and absorption. Neutron methods are neutron absorption, PGAA and INAA.

A number of review papers summarizing general applications of PGAA have been published.[23-25] By far, the most common use today of PGAA analyzers is in on-line measurements of coal quality. These include monitoring of product quality, blending to specifications, sorting and so on. For the principal elements in coal, hydrogen and iron it provides a good response. Carbon in coal responds poorly, however, its concentration is high enough to be detected. The quantities of most interest are ash, sulfur, moisture, and metalliferous minerals. The characteristic prompt gamma-rays from the elements of interest in coal analysis are summarized in *Table* 3. The determination of C, H, N, S, Al, Si, Ca, Ti, Fe, Na, K, and Cl in coal or lignite is possible with high precision and accuracy within 15 minutes using ^{252}Cf with a source strength of about 200 μg and a HPGe detector. The analysis of a raw glass mix for Na, Si and Ca could be achieved within 6 minutes.[26]

Because of the low detection limits achievable for a number of elements, the PGAA method is used in solid state physics, analytical chemistry, biology, metallurgy and medicine. PGAA is also one of the useful methods for the non-destructive measurement of low Z elements for which the neutron capture does not produce radioactive

Table 3. Prompt gamma-ray elements influencing coal quality

Parameters of coal quality	Elements	Neutron reaction	Prompt γ-ray energy (keV)
Combustion heat	C	C(n, n'γ)	4443
	O	O(n, n'γ)	6129
Ash content	Al	Al(n, γ)	7724
		Al(n, n'γ)	844, 2210
	Fe	Fe(n, γ)	7632, 7645
		Fe(n, n'γ)	846
	Ca	Ca(n, γ)	1943, 6410
		Ca(n, n'γ)	3736, 3904
	Si	Si(n, γ)	3539, 4934
		Si(n, n'γ)	1779
Moisture	H	H(n, γ)	2223
	C	C(n, n'γ)	4443
Sulfur Content	S	S(n, γ)	841, 2380, 5420

nuclides, such as hydrogen, boron, and nitrogen. Thermal neutron has been used for the determination of hydrogen, usually at a mass fraction of above 1%, in food. For the determination of hydrogen concentration below 1%, cold neutron can be used.[27] Thermal or cold neutron PGAA is very useful for the determination of boron, since it is difficult to measure by other conventional analytical techniques. Boron in various materials such as steels, polymers, foods or meteorites has been measured at lower ppm levels using cold neutron.[28] Kjeldahl method is normally used for the measurement of nitrogen, but does not always yield total nitrogen. A total nitrogen mass fraction of 1% or higher can be measured by thermal neutron PGAA and cold neutron PGAA for the nitrogen mass fraction below 1 %.

Measurement of the unique prompt γ-rays from nitrogen can be applied to the detection of explosive materials, since the nitrogen is a major element in explosives. Such a PGAA technique is a newly proposed alternative to screen suspected materials in baggage for airport security.[29] In this system, the baggage moves through a screen of thermal neutrons produced by a ^{252}Cf source; the detected 10.83-MeV gamma-rays are tomographically analyzed to give the spatial distribution of nitrogen. Chung et al. used the high-density 2″×2″ BGO detector and a 20 μg ^{252}Cf neutron source in order to increase the counting efficiency, which makes the PGAA explosive detection system more effective for practical use.[30]

A series of other applications are possible besides these applications mentioned above, since almost all chemical elements emit some characteristic γ-rays after neutron capture. Medical application of PGAA of course depends on the same principles as the non-medical applications mentioned earlier. A PGAA system using a neutron beam from the 30MW research reactor at HANARO, KAERI was installed to determine boron content in biological samples for the boron neutron capture therapy (BNCT) using a single n-type HPGe detector with a thermal neutron flux of 6.1 x 10^7 $n \cdot cm^{-2}s^{-1}$.[31,32] The BNCT uses ^{10}B(n, α)^{7}Li reaction from which alpha particles are emitted and therefore a tumor cell could be cured selectively. Since fast and accurate quantitative analysis of ^{10}B is required prior to therapy in order to determine the neutron irradiation time for BNCT, the PGAA can be considered as a very suitable method.

6. On-going and future program in KAERI

A device using the principle of PGAA has emerged as an important analytical tool, since it can provide an accurate, fast, automated on- or in-line measurement. The development of new facilities for application to various fields as well as the improvement of the current PGAA will be of great benefit to scientists worldwide.

The PGAA system using a ^{252}Cf source at the Korea Atomic Energy Research Institute is currently under construction for the on-line measurement of several elements in aqueous samples.[33] For background suppression, the γ-γ coincidence method is utilized to improve the detection limit. Four-π geometries for the neutron source are applied to enhance the sensitivity of detection. Neutron reflecting materials such as pyrolitic

graphite or carbon can be used around the sample container to enhance the effectiveness of neutron irradiation. The PGAA technique could be used for analyzing a wide variety of different materials and is especially very useful for the detection of chemical weapons or explosives as well as various narcotics, since the ^{252}Cf neutron source could possibly make the system portable.

7. Conclusion

PGAA is a rapidly developing analytical technique applicable to all elements including light elements, which are usually difficult or even impossible to measure by conventional analytical tools. It is a complementary analytical tool for delayed neutron activation analysis. PGAA has various advantages and disadvantages compared to the conventional delayed NAA. The advantages include rapid analysis with high accuracy and the ability to measure elements, which have no suitable isotope for delayed NAA. On the other hand, the neutron fluxes used are lower and thus give lower sensitivity. Another disadvantage is that the spectra are very complex and detectors are subject to damage from neutron irradiation. However, using the γ-γ coincidence method can improve the peak to background ratio substantially, therefore improves the detection limit.

Application of PGAA technique to various industries for on-line analysis of coal and mineral ore as well as explosives have proven to be an attractive alternative to the conventional methods. The future of the PGAA is strongly dependent on the reliability, operating costs and the possibility of use in other industries. Despite the many advantages of PGAA, a series of problems have to be solved in order to have a broader application. The counting conditions, system geometry, shielding design and coincidence detection system have to be optimized or established to meet the requirements of specific measurement.

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