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Nuclear Instruments and Methods in Physics Research A 566 (2006) 442–447
NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH
Section A
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Analytical capability of an explosives detection by a prompt gamma-ray neutron activation analysis

Hee-Jung Im[a,*], Hyun-Je Cho[b], Byoung Chul Song[a], Yong Joon Park[a],
Yong-Sam Chung[c], Won-Ho Kim[a]

[a]Nuclear Chemistry Research Division, Korea Atomic Energy Research Institute, 150 Deokjin-dong, Yuseong-gu, Daejeon 305 353, Republic of Korea
[b]Nuclear Environment Technology Institute, Korea Hydro & Nuclear Power Co. LTD, 150 Deokjin-dong, Yuseong-gu, Daejeon 305 353, Republic of Korea
[c]HANARO Utilization Technology Division, Korea Atomic Energy Research Institute, 150 Deokjin-dong, Yuseong-gu, Daejeon 305 353, Republic of Korea

Received 9 May 2006; received in revised form 14 June 2006; accepted 15 June 2006
Available online 14 July 2006

Abstract

Neutron-induced prompt gamma-ray analysis method was evaluated for the detection of explosives by measuring the H, C(C/H), and N(N/H) concentrations of samples at a research nuclear reactor.

The hydrogen concentration was measured from the calibration curves obtained from various samples which contained hydrogen, and the detection limit for the determination of hydrogen in the samples was estimated. Standard reference materials (NIST SRMs) were used as analytical controls.

The γ-spectra obtained here will be directly used for further studies as a training set for a pattern recognition to develop suitable discriminant classes with which explosives can be distinguished from innocuous materials.

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PACS: 07.85.Nc

Keywords: Prompt gamma-ray neutron activation analysis (PGNAA); Thermal neutron; Explosive detection; Hydrogen; Elemental composition

1. Introduction

Detection of explosives can be achieved based on certain parameters such as the geometry, vapor emissions and elemental compositions of the samples of interest. The shape of the explosives can be detected with an X-ray imaging system. Since the geometry of explosives can be modified into various shapes like toys or foods to hide the presence of a suspicious object, the presence of a metallic detonating system has to be detected instead. This method contains difficulties for inspectors in that finding such a small wire in a detonating system even with a high-resolution X-ray system is not an easy task. The second approach is the use of a so-called vapor sniffer which measures any traces of the characteristic volatile compounds from explosives. However, this method requires the withdrawal of a sample from the enclosure, and there are interferants for an explosive vapor detector such as dirt, tobacco smoke, cigarette ash, perfumes, body odors, and so on, even though such a system is relatively sensitive. The final approach involves a detection of the major constituents of explosives such as N, O, C, H and Cl [1].

In recent years, considerable effort has been directed toward detecting the elemental compositions of explosives by means of a thermal neutron interrogation, which involves exposing baggage to slow neutrons with an energy level in the order of 0.025 eV [2]. An explosive agent is composed primarily of inorganic nitrates and carbonaceous fuels. Although a certain concentration level of nitrogen is a good indicator of explosives, the combinations of oxygen, hydrogen and carbon should be considered since some common compounds such as melamine and silk contain high concentrations of nitrogen similar to

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*Corresponding author.
E-mail address: imhj@kaeri.re.kr (H.-J. Im).

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doi:10.1016/j.nima.2006.06.044

explosives. A measurement of the carbon and oxygen, and nitrogen and oxygen densities (mol/cm^3) respectively, provides for a good separation of explosives from non-explosives [3].

Since neutrons and γ-rays are highly penetrative for most materials, an analysis using these can be performed non-destructively and in-situ, thus removing all questions of an incomplete dissolution, contamination, size and shape dependency, and a loss of samples. By using the prompt gamma-ray neutron activation analysis (PGNAA) method, not only fissile materials (such as plutonium-239 and californium-252), but also trace elements (Cd, Sm, and Gd) which have large cross-sections and light elements (H, B, C, N, Si, P, S, and Cl) can be determined [4] while the neutron detection methods [5,6] can only detect fissile materials which emit neutrons via spontaneous fissions. In a PGNAA, a target is placed in a beam of neutrons. The γ-rays being emitted upon a neutron capture in the target are measured by a shielded germanium detector, and they yield quantitative elemental peaks. We have used a radioisotope neutron source [7], and we are setting up an accelerator-based neutron source [8] created from a deuterium–deuterium (D–D) reaction in our lab in cooperation with a group from the Lawrence Berkley National Laboratory (LBNL) because of the advantages of their size and compactness. For a better detection of the neutron or γ-rays, a high flux, a good scintillator [5,6] or an effective noise reduction [9] is necessary, and here, we use a neutron beam from a research reactor for the best sensitivity [10]. In the earlier papers [11,12], the elemental sensitivities and the detection limits especially for boron and a few other elements were obtained by using the PGNAA facility at the 24 MW HANARO research reactor of the Korea Atomic Energy Research Institute (KAERI), but the ones for hydrogen have not been done as yet.

A single hydrogen peak can be easily measured because its signal is independent of the chemical form of the hydrogen present, but since all of the materials used in the PGNAA setup produce prompt γ-rays by the capture of scattered neutrons from the sample and the sample holder, a careful analysis of the background in the PGNAA becomes inevitable. The present study is to analyze the hydrogen concentration of the used samples by using a calibration curve and to measure the N/H and C/H concentrations to demonstrate the capability of this method for detecting explosives from among non-explosives. The count rates of the samples by a Compton suppression, single-mode and DSPEC are investigated, and the standard reference materials are irradiated for an analysis assurance.

2. Experimental

2.1. PGNAA system

Present study was implemented by using the PGNAA facility at the 30 MW HANARO research reactor of KAERI. The thermal neutron PGNAA facility is located about 4 m above the reactor core, and the diffracted beam passes through a 1 m long collimator to the PGNAA beam shutter. A sample is irradiated in a beam size of $2\times2\,\mathrm{cm}^2$ with a neutron flux of about $1.4\times10^8\,\mathrm{n\,cm^{-2}\,s^{-1}}$. The detector system consists of a high-purity germanium (HPGe) detector surrounded by eight bismuth germanate (BGO) and two thallium-doped sodium iodide (NaI(Tl)) scintillators as an annulus type to reject the Compton scattered photons as demonstrated in a previous paper [12]. An HPGe detector (43% efficiency relative to a $7.6\,\mathrm{cm}\times7.6\,\mathrm{cm}$ sodium iodide crystal, EG&G Ortec, USA) connected to a computer-based 16 k channel MCA (919 MCB, EG&G Ortec, USA) or DSPEC (EG&G Ortec, USA) is used for counting the prompt γ-rays. The resolution of the detector is 2.2 keV at 1332 keV ^{60}Co. The sample is usually positioned 25 cm away from the face of the detector. A Teflon sheet is used to suppress the background appearing from a sample's casing. The variations in the neutron flux are monitored by a periodic irradiation of a Ti foil.

2.2. Sample preparation

The powdered samples were dried by using an oven at 30 °C for 2 h and then cooled at room temperature. The prepared samples were put into Teflon vials with lids and stored in a desiccator. To check on the moisture content, the samples were weighed before and after being dried. The moisture content of the sample was less than 0.15% from the weight difference after being dried. The weight of a sample was measured three times per each sample. The chemicals used in this study are the following; melamine (MEL, $C_3H_6N_6$, Aldrich, 99%), 4-nitrophenol (NP, $C_6H_5NO_3$, Fluka, 97%), p-aminobenzoic acid (ABA, $C_7H_7NO_2$, Fisher, 98%), nitrobenzene (NB, $C_6H_5NO_2$, Osaka, 99%), triethyl phosphate (TP, $C_6H_{15}O_4P$, Yakuri, 99%), polyethylene (PE, $(\text{–}CH_2CH_2\text{–})_n$, Aldrich), sodium nitrite ($NaNO_2$, Junsei, 99%), sodium nitrate ($NaNO_3$, Kanto, 99.95%), and sodium carbonate (Na_2CO_3, Aldrich, 99.95%). Samples of MEL (1.1, 2.6, 5.7, 16.2, 46.7, 101.81 and 153.5 mg), NP (101.93 mg), ABA (101.55 mg), NB (107.88 mg), PE (12.3, 20.8, 56.8, 123.3 and 161.7 mg), TP (121.36 mg) and mannitol ($C_6H_{14}O_6$; 10.3, 20.7, 40.3, 80.8 and 100.3 mg) were placed in Teflon vials to be tested. Imitators of ethylene glycol dinitrate (EGDN, $C_2H_4N_2O_6$) and pentaerythritol tetranitrate (PETN, $C_5H_8N_4O_{12}$) reproducing the same relative concentrations of carbon, hydrogen, nitrogen, and oxygen as in the real ones were prepared from a mixture of PE, $NaNO_2$, $NaNO_3$ and $NaCO_3$. Sample A ($C_3H_6N_3O_6$) was also prepared by the same method as the imitators.

The standard reference materials (NIST SRMs) of 1632c, 1566b, 1573a, 1570 and 1549 were also used for an analytical control purpose.

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