Intercomparison and determination of environmental standard samples by instrumental neutron activation analysis

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To identify and improve the analytical technique for air pollution research, four kinds of environmental standard samples, i.e., airborne particulate matter, coal fly ash, soil and pine needle supplied from the NIST and the IAEA were analyzed using thermal and epithermal neutron activation techniques. Sample irradiation was done at the irradiation facilities (neutron flux, \(1 \times 10^{13} \text{n} \cdot \text{cm}^{-2} \cdot \text{s}^{-1}\)) of the TRIGA MARK-III Research Reactor in the Korea Atomic Energy Research Institute. The accuracy and precision for the analysis of 40 trace and toxic elements in the samples were compared with the certified and reported values, respectively. In the analytical results of all standard reference materials, the relative standard deviation were within the 15% except for 11 elements and the relative error were agreed within the 10-20% except for 13 elements. The benefit of epithermal activation was investigated and the optimum analytical condition is reported.

Introduction

Environmental conditions are changing rapidly these days in a global scale. In view of the increasing concern both at home and abroad regarding environmental problems, it is important to measure elemental concentrations in environmental samples such as aerosol, earth's crust, water and other biological samples, etc. for environmental monitoring and air pollution study. Instrumental neutron activation analysis (INAA) with the recent development of high performance γ-ray counting system has become one of the most powerful methods in nuclear analytical techniques (NAT) for the non-destructive, simultaneous multi-element analysis of trace level component in various samples. The capability of rapid and accurate analysis of many samples with the help of computer and automatization makes INAA of wide application. Particularly, it has a good advantage in analysis of environmental samples such as airborne particulate matters in atmosphere consist of various trace elements. Nowadays, this method is also recommended and used for the standardization of analytical techniques internationally. In the near future, routine analysis will be carried out speedily with the development of rapid, accurate and automatic analytical methods for these materials. The results can be used either in undertaking background monitoring; to conduct epidemiological studies, to investigate source apportionment methodologies and source–receptor models, to study long-range transport phenomena, and to establish a more cost-effective plan to control pollution.

In this study, non-destructive thermal and epithermal neutron activation analysis of the component elements in four kinds of environmental samples have been investigated. In addition, the three IAEA's samples were analyzed for the standardization of analytical procedure and data intercomparison according to international co-operative research.

Experimental

Facilities and equipments

TRIGA Mark-III Research Reactor, KAERI: PTS, RSR and Cd lined irradiation facilities (neutron flux \(1 \times 10^{13} \text{n} \cdot \text{cm}^{-2} \cdot \text{s}^{-1}\)).

Gamma-ray spectrometer: HP Ge Semiconductor Detector, EG&G ORTEC; GMX series (3 KeV–10 MeV), GEM Series (50 KeV–10 MeV); 25% relative efficiency, 1.9 KeV resolution at 1332 KeV of 60Co; peak to compton ratio: 45 : 1.

Low background Pb shield for Ge detector, EG & G ORTEC: 4π–10 cm thick low background virgin lead, graded Cu & Cd lined, 28 × 41 cm² cavity;

LN₂ automatic transfer system; multichannel analyser, EG & G ORTEC: 918A MCB; 8K channel ADC (10 μs), 8K data memory, counting loss correction; 919A MCB; 16K channel ADC (7 μs), 64K data memory, digital stabilizer MAESTRO-II emulation software; application software for NAA: gamma-ray spectrum analysis software.

MINIGAM: data acquisition and analysis, on-line, EG & G ORTEC, USA.

OMNIGAM: advanced data acquisition and analysis, wide variety of spectral correction routine, on-line, EG & G ORTEC, USA.

NAA KAERI: private analysis program for NAA.

Sample and sample preparation

Standard reference materials: To assess the analytical processes and data intercomparison, four kinds of standard reference materials (urban particulate matter SRM 1648, coal fly ash SRM 1633a, soil SRM 2709 and pine needle SRM 1575, the National Institute of Standards and Technology, NIST) were chosen as common environmental samples. Standard samples of fine powder were pre-treated with the recommended method in NIST certificate. The samples were dried for 8 hours at 105 °C.
and then cooled at room temperature in the desiccator. Samples of 10–200 mg were weighed and put into polyethylene vials and heat sealed. The high purity polyethylene sample vials were pre-washed with spectrograde dilute nitric acid and acetone, and the blank test performed to identify the impurity concentrations.

IAEA’s samples: Three kinds of environmental samples, urban dust (IAEA 395), coal fly ash (IAEA 394), soil (IAEA 327N) are supplied by the IAEA. Samples are pre-treated as mentioned above and about 10–200 mg were weighed accurately and put into polyethylene vial and heat sealed.

Irradiation and gamma-ray counting

Samples were irradiated with thermal neutrons from TRIGA Mark-III Research Reactor in the Korea Atomic Energy Research Institute. Pneumatic Transfer System (PTS, \( \Phi_T = 1.2 \cdot 10^{13} \text{n} \cdot \text{cm}^{-2} \cdot \text{s}^{-1} \)) was used for the short time irradiation and Rotary Specimen Rack (RSR, \( \Phi_T = 3.0 \cdot 10^{12} \text{n} \cdot \text{cm}^{-2} \cdot \text{s}^{-1} \)) for the medium and long time irradiation. Epithermal neutron irradiation was carried out in Cd tube (CdT, \( \Phi_T = 5.0 \cdot 10^{10} \text{n} \cdot \text{cm}^{-2} \cdot \text{s}^{-1} \)) which is lined with 0.5 mm cadmium plate. Samples were irradiated at the same geometric position to minimize the geometry error. For neutron flux monitoring, activation wire (Reactor Exp. Inc., R/X activation wire, 99.99% purity) such as Au–Al, Cu, and Mn were used. The analytical condition was optimized after considering the preliminary results. Energy and efficiency calibration were done with the radioactive multi-nuclide reference source (NEN Products Inc., NES-602, 1" diameter disc type) certified by NIST. Dead-time of detector was maintained under 5% as much as possible. Decay and pile up correction were done automatically. Instrumental and analytical condition must be set with the extra efforts in the case of the analysis.