Enhancements and Health-Related Studies of Neutron Activation Analysis Technique

A Thesis Submitted
By

Mohamed Ali Moawad Soliman
Atomic Energy Authority

To

Department of Chemistry,
Faculty of Science,
University of Ain Shams

In
Partial Fulfillment of the Requirements for a
PhD Degree in Chemistry

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M. Soliman

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Abstract

The work presented in this thesis covers two major points. One algorithm concerns with establishment of an accurate standardization method with multi-elemental capabilities and low workload suitable for NAA standardization at ETRR-2. The second one deals with constructing and developing an effective non-destructive technique for analysis of liquid samples based on NAA using (very) short-lived radionuclides.

To achieve the first goal, attention has been directed toward implementation of the k_0-method for calculation of the elements concentrations in the samples. The k_0-method of NAA-standardization has a considerable success as a method for accurate multi-elemental analysis with comparable low workload. The k_0-method is based on the fact that the unknown sample is irradiated with only one standard element as comparator. To access the implementation of this method at ETRR-2, careful and complete characterization of the neutron flux parameters in the irradiation positions as well as the efficiency calibration of the γ-ray spectrometer must be carried out. The required neutron flux parameters are: the ratio of the thermal to epithermal neutron fluxes (f) and the deviation factor (α) of the epithermal neutron flux from the ideal 1/E law. The work presented in Chapter 4 shows the efficiency calibration curve of the γ-ray spectrometer system which was obtained using standard radioactive point sources. Moreover, the f and α parameters were determined in some selected irradiation sites using sets of Zr-Au as neutron flux monitors. Due to different locations relative to the reactor core, the available neutron fluxes in the selected irradiation positions differ substantially, so that

different irradiation demands can be satisfied. The reference materials coal NIST 1632c and IAEA-Soil 7 were analyzed for data validation and good agreement between the experimental values and the certified values was obtained. The obtained results have revealed that the k_0-NAA procedure established at the ETRR-2 can be regarded as a reliable standardization method of NAA and as available analytical method for elemental analysis of samples especially those for which are difficult to find a proper reference material. The analysis of reference materials indicates that under our experimental conditions, results may have maximum biases of less than 5% from the true values for elements analyzed using the so-called 1/v nuclei.

To solve the problems of liquid samples, *Flowing Sample Neutron Activation Analysis (FSNAA)* set-up was constructed in this work. The developed set-up involves a continuously flowing of the liquid sample between the irradiation site and the detector in a polyethylene tube line by the aid of mechanical pump. Due to unavailability of irradiation facility in ETRR-2 suitable for accommodation of the developed set-up, a ^{252}Cf neutron source was used to carry out the irradiation process. This arrangement aims to achieve several objects. Firstly, the complicated pretreatment (preconcentration) steps of liquid samples can be omitted via analysis of large volume of the sample. Secondly, saving time and irradiation containers since the preparing of replicates is not required. Finally, the effect of dead time can be minimized, since flowing sample continuously feeds the detector with fresh radioactive material, keeping the radioactivity level constant during the whole period of measurement.

Modeling calculations using MCNP, version 5 were performed to optimize the γ-ray counting configurations and derive the required correction factors. An MCNP simulation was used to define the best thickness of paraffin fixed between the source and helical irradiation hose to get the highest intensity of thermal neutrons.

The repeatability of FSNAA was investigated. Less than 3% standard deviation between the results of several measurements was achieved. The detection limits, accuracy and ability to remove spectral interference of the new system were tested by analysis of synthetic single and multi-elements standards solutions. The obtained detection limits compare favorably with those obtained by conventional INAA methods and other analytical method. Besides, the developed method seems to have many advantages when compared with conventional INAA methods. These include:

- Ability to analyze liquid samples without preconcentration steps
- Low detection limits
- Simplicity and no need for blank correction
- Ability to analyze inhomogeneous solutions (like colloidal or suspended solutions) with pretreatment processes, if total elemental content is required

In addition, the developed FSNAA procedure was tested for determination of trace and ultra-trace elements of nutritional and toxicological significance in water sample collected from the Mediterranean Sea in some areas of Alexandria, Egypt. Therefore, it is necessary to determine its elemental content in order to

evaluate its effect on food chains and hence on the health of human being. Due to low intensity of the neutron source, only Na and Cl were determined in the examined sample. It was expected that more elements could be measured if high neutron flux source (like nuclear reactor) is used.

The obtained results indicate that the FSNAA is a simple procedures which is promising analytical method for analysis of liquid samples with low detection limits. It can be used for monitoring the elements levels in environmental samples (water or compressed air), industrial wastewater and other liquid samples. It can also be utilized for analysis of urine and other animals' effluents for health-related studies.

Contents

List of Symbols

k'	Calibration constant
ϕ_{ep}	Epithermal neutron flux of energies between 0.55 e V and 0.5 MeV
ϕ_f	Fast neutron flux for energies more than 0.5 MeV
σ_r	Method repeatability
ϕ_{th}	Thermal neutron flux for energies up to 0.55 eV
ζ	Zeta score
$\phi(E)$	Neutron flux (neutron.cm^{-2}.s^{-1}) of neutron with energy E
A	Source strength
A_p	Full-energy peak count rate
b	Barn, unit of cross-section. 1 b = 10^{-24} cm^2
C	Counting factor
D	Decay factor
D_L	Detection limit
E	Neutron energy
f	Thermal neutron to epithermal neutron fluxes ratio
I_0	Infinite dilution resonance integral
k	k-factor
k_0	k_0-factor
M	Atomic weight
N	Number of radioactive nuclei