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PROMPT GAMMA NEUTRON ACTIVATION ANALYSIS (PGNAA) IN NUCLEAR TECHNOLOGY

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Abstract

The scope of the present thesis has been to develop and standardise suitable analytical methodology for determination of trace elements particularly boron and cesium, both being important from their relevance in nuclear technology. The former is important in the front-end of nuclear fuel cycle while the latter is of relevance in the back-end of the nuclear fuel cycle. In view of the requirement of boron determination in a wide variety of matrices, the investigations were undertaken to adapt as many suitable techniques as possible for different matrices. At the same time the possibility of employing more than one technique for validation of analytical results was also emphasised. Final acceptance of any analytical result is decided by the statistical treatment to which the data is subjected to. ISO recommended total measurement uncertainty principle has been adopted. Prompt Gamma Neutron Activation Analysis (PGNAA) is a nuclear analytical technique for the analysis of elements present in solid, liquid and gaseous samples by measuring the capture gamma rays emitted from the samples during neutron irradiation.

Discussion and Result: PGNAA is one of the sensitive methods to determine boron in various matrices. [1] Since it is a matrix independent technique, matrix matched standards are not essential for each and every analysis. Moreover, it's a one of the preferable methods as a reference for certifying the value in any standard. Capture gamma rays also called prompt gamma rays since they are emitted within about 10-12 s of capture of neutron by an atomic nucleus. So neutron irradiation and gamma ray counting of the sample is done simultaneously in PGNAA. It is therefore an online technique and preferably requires a "neutron beam" facility for irradiation of targets. On the average 3-4 γ rays are emitted per neutron capture and they carry off about 7-10 MeV excitation energy, which is the average binding energy of a neutron in a nucleus. The prompt gamma rays can therefore have energies in the range of a few keV to about 10 MeV. Prompt gamma rays are characteristic of the isotopes of elements and practically all isotopes of all elements absorb neutron and emit prompt gamma rays. Hence, all elements in entire periodic table can in principle be

measured employing prompt gamma rays. However, it is one of the suitable methods to determine low atomic number elements like H, B, C, N, Si, P, S and Cl and elements like Hg, Cd, Sm and Gd which are having large neutron absorption cross section. The main advantages of this technique are i) non-destructive multi-elemental bulk analysis is possible which offers the flexibility of sample size and shape and can be particularly suitable for archeological, geo and cosmo-chemical analysis, ii) because of higher γ ray energy there is a minimum attenuation in the sample and hence larger sample size can be used for analysis. The main limitations of this methodology are (i) the prompt gamma ray spectra are very complex, often containing several hundred peaks necessitating the use of a suitable peak fitting software, (ii) a separate neutron beam facility is required, (iii) existing prompt gamma ray data are inadequate and (iv) sensitivities for most of the elements are lower than the conventional neutron activation analysis. As mentioned above, dedicated neutron beam facility is required for PGNAA. In general neutron beam lines used for prompt gamma experiments can be broadly classified.

Guided Beam : Here neutron beam is transported from reactor core to experimental site through a beam tube. This offers the following advantages: (1) low stray neutron and gamma ray background (2) both thermal and cold neutron beam can be used (3) minimal interference from fast neutrons due to resonance. Cold neutron beams offer higher sensitivity due to higher beam intensity and enhanced absorption cross section arising from 1/v law.

Diffracted Beam: Neutron beam coming out of reactor core is reflected by a suitable crystal and taken to the experimental site. In this case also high gamma ray background of the radial neutron beam is avoided. Here is beam is composed of neutrons having selective wavelength. **Spallation Neutrons:** Neutrons emitted from a spallation reaction in a cyclotron are thermalised and used as a beam. The characteristic of such beam is that there is no gamma ray background associated with a reactor and higher neutron beam intensity is also realised.

Shielding requirements: Since stray neutron and gamma ray background are usually very high around a neutron beam line, detector shielding is an important aspect of PGNAA set up. Gamma ray background is reduced using a thick lead shield while neutron absorbers like Cd and B can be used for neutron shielding. However, B and Cd produce 478 keV and 559 keV (and higher energy) γ rays respectively on neutron absorption which contribute to the background. Neutrons also react with detector cover cap and Ge to produce ambient gamma ray background. Hence enriched 6Li in the form of LiF ceramic tile or in other form is

commonly used as cover cap of the detector to prevent neutron from entering the detector. 6Li (n, α) 3T reaction does not produce any γ ray.

Target preparation and irradiation: Normally solid targets in the range of 0.1 to 1.0 g in powder, pellet or disc are used for irradiation. For liquid samples upto 2 mL could be taken and also it depends on the volume of sample holder and neutron beam size. Size of the target should be less than the neutron beam size. If comparison with a standard is done, then errors due to matrix effect (neutron self shielding, neutron scattering, gamma ray attenuation and change in detector efficiency due to sample geometry) can be minimized by matching geometric configurations of sample matrix as closely as possible to that of standard. For irradiation, samples are usually wrapped or sealed into Teflon bags and suspended in the neutron beam using Teflonstring. Teflon produces minimum background since both carbon and fluorine have low neutron absorption cross section (in the order of mb).

Instrumentation: The instrument used for PGNAA should be a high resolution, high efficiency γ ray spectrometer system. Generally high energy γ rays will be emitted in PGNAA analysis. Hence, a large volume high purity germanium detector is required to meet the requirement of PGNAA analysis. In order to reduce the Compton background in the spectra, the HPGe detector is often used with an anti Compton annular BGO (bismuth germanate) shield which surrounds the HPGe detector. Since a Compton scattered gamma ray from HPGe detector is intercepted by the anti Compton shield, the signal from HPGe detector is rejected if it is associated with a signal from the shield.[2,3] Thus Compton background could be appreciably reduced, which in turn improves the detection limit of measurement. Since, pair production becomes the primary mode of interaction for high energy γ rays in the detector material, higher energy portion of the prompt gamma spectrum is complicated by the presence of single and double escape peaks in addition to photo peaks. Thus every γ ray produces three peaks. In order to reduce the complexity, the anti Compton shield is also used in pair spectrometer mode. In this case, the coincidence signal from the two segmented halves of the shield in association with HPGe detector signal is registered as valid event. Thus the higher energy portion of the spectra becomes free from the photo peak and single escape peak, thereby making the spectra simpler. In practice, data acquisition is simultaneously carried out in singles, anti Compton and pair mode and sorted out during data analysis time. Data acquisition is done in a pulse height analyser (PHA) with 8k memory.

Methodology: The count rate R (cps) due to ith isotope of an element present in the sample is given as R (cps) = WθiNoσiφaiεi/M ..... (1) Where W is weight of the element in the

sample, and M is its atomic weight. θi, σi and ai are the isotopic abundance, cross section and gamma ray abundance of the ith isotope of the element of interest. εi is efficiency of detection of gamma ray of interest. No and φ are Avogadro number and flux of the neutron beam respectively. As evident from the equation above, the detection sensitivity of an element is given by the product (θiσiφaiεi) and higher value of any one of the parameters enhances the sensitivity of detection.

Radioactivity counting techniques based on gamma spectrometry offer a very sensitive measurement option. However, these involve expensive detection and data analysis systems. Moreover these can be applied only for measuring radioactive cesium. It would be ideal to have a measurement technique, which is reasonably sensitive, easily portable and at the same time can also be applicable to measurement of stable cesium in environment.

The highlights of the present investigations are:

(1). Using low neutron fluxes, about 106 n/cm2/Sec, boron in ppm levels was determined employing Chemical PGNAA. This is the first time chemical PGNAA has been employed to determine boron. (2). The well-known spectrophotometric method for the determination of boron in uranium compounds with curcumin after extraction with 2-ethyl hexane 1,3 diol has been modified such that the sample size is reduced from 2.5g to 0.200g. Extensive investigations have been carried out to study the reasons for nitric acid interference in methodology and to circumvent the same. These studies are essential in nuclear technology since different acids are employed to dissolve uranium compounds. (3). To our best of knowledge, first time spectrophotometric method has been developed and employed for the determination of boron in uranium-aluminum-silicon inter metallic compound. This method has been extended to determine boron in various silicon related materials of interest in nuclear technology like SILUMIN, Al-Si alloy and U3Si2. (4). Spectrophotometric method employing curcumin has been modified for the determination of boron in highly refractory material Zr-Nb alloy. Dissolution procedure has been modified suitably to prevent the loss of boron. Due to selective separation of boron with 2-ethyl hexane 1,3 diol, reported interference from tin was avoided and also precision and accuracy has been improved considerably compared with literature reports. (5). To increase the confidence on measurand values, uncertainty in spectrophotometric method for the determination of boron was estimated by following ISO guidelines.

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