

Neutron Activation Analysis: A Versatile Nuclear Analytical Technique for Chemical Characterization of Materials

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Abstract

Neutron Activation Analysis (NAA) is a versatile nuclear analytical technique for quantification of elemental concentrations simultaneously at major, minor and trace levels in a wide variety of matrices with good accuracy and precision. Conventional NAA in combination with Prompt gamma-ray NAA (PGNAA) can determine as many as 70 elements starting from Hydrogen to Uranium. Principles and capabilities of PGNAA, conventional NAA and fast (neutron) NAA (FNAA) are described briefly. Since NAA utilizes neutrons and gamma-rays, it experiences less or negligible matrix effect and thus it is capable of analyzing small (a few mg to less than a gram) as well as large size (kg scale) samples. Instrumental NAA (INAA) is most common approach for quantifying many elements, however depending on matrix of sample, pre or post irradiation chemical separation known as preconcentration NAA (PNAA) or radiochemical NAA (RNAA), respectively, is carried out in order to determine elements of interest. The elemental concentrations can be determined by relative method using elemental standards or by single comparator method known as k_0 -based NAA. Conventional and k_0 -NAA methods are validated by analyzing certified reference materials (CRMs) as a part of quality control (QC) exercise. NAA methods have been applied in many fields of

science and technology like materials science (metals, alloys, glass and ceramics), high purity materials, geology, ores and minerals, environment, biology, food and agriculture, industry, archeology and forensic sciences. NAA methods were used for determination of total elemental concentration as well as speciation of elements like As, I and Cr and bio-accessibility of trace elements in edible plants and selenium in food samples. Many trace elements could be determined due to the use of high thermal neutron flux (10^{11} - 10^{14} n cm⁻²s⁻¹) from research reactors in conjunction with high efficiency and high-resolution gamma-ray spectrometers using HPGe detectors.

Keywords: NAA, PGNAA, FNAA, k_0 -NAA, Multielements, Materials Science, Geology, Environmental, Biological, Food, Agriculture, Industry, Archaeology, Forensic Science

1. Introduction

Neutron Activation Analysis (NAA), a branch of activation analysis, is a versatile technique for determination of large number of elements in wide varieties of matrices. NAA is a sensitive and selective analytical technique capable of giving results of many elements even from a small amount (a few mg level) [1,2]. George de Hevesy and Hilde Levi were first to introduce the NAA technique in 1936 [3]. By varying parameters like neutron flux, duration of irradiation, cooling time and using both instrumental and radiochemical techniques in NAA, it is possible to accurately determine many elements. Currently, NAA method is being widely used for multi-elemental determination in a variety of matrices like geological, biological, environmental, archaeological, precious materials (gemstones), high purity and reactor structural materials [4-7]. The high sensitivity of this technique is because of irradiation of samples at high neutron flux available from the research reactors and measurement of gamma rays from the samples using high-resolution high purity Germanium (HPGe) detector coupled to multichannel analyzer (MCA). The technique provides a possibility to perform analysis non-destructively using instrumental neutron activation analysis (INAA). Many elements from Na to U including transition elements and rare earth elements (REEs) are routinely determined by NAA methods.

1.1 General considerations

NAA is based on nuclear reaction phenomenon. When a neutron interacts with a target nucleus, a compound nucleus is formed. The compound nucleus has a certain finite lifetime ($\sim 10^{-13}$ s). De-excitation of the compound nucleus can occur in different ways that are independent of the way the compound nucleus is formed. Each of these processes has a certain probability, depending on the nuclear cross-section of each mode. The most favorable and important nuclear reaction in NAA is radiative capture (n,γ), in which the excited nucleus passes to a lower energy state by the emission of one or more γ -rays, called prompt gamma-rays. The resulting nuclide is usually radioactive. A radioactive nuclide has a characteristic half-life ($t_{1/2}$), mode of decay and energy of emitted radiation(s) during the decay process, which are like finger prints. Depending on the energy considerations, a radionuclide can decay to a daughter product by various ways. In general, the majority of the radionuclides formed by the radiative capture (n,γ) process undergo beta decay, and in most cases beta decay is associated with emission of one or more gamma rays. In fact, a very few radionuclides undergo pure beta decay (^3H , ^{14}C , ^{32}P , ^{35}S etc.). Positron decay and electron capture are favorable in radionuclides that are neutron deficient.

1.2 Principle of NAA

In NAA, neutron is used as the probe. Interaction of neutron with the target nucleus results into the formation of compound nucleus in an excited state. Binding energy of the thermal neutron with the nucleus governs the excitation energy of the compound nucleus. Prompt gamma-rays (neutron capture gamma-rays) are emitted from the compound nucleus instantaneously to a more stable configuration (activation product). The activation product, in many cases, de-excites by emission of one or more characteristic delayed gamma rays, following the unique half-life of the radioactive nucleus. Depending upon the particular radioactive species, half-lives can range from very short (fractions of a second) to very long (several years). The different processes of NAA are shown pictorially in Fig. 1.

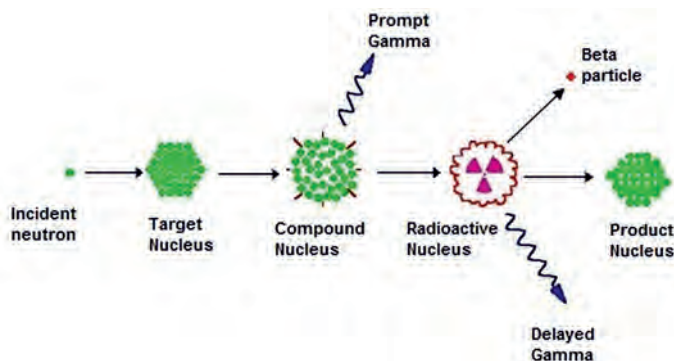


Figure 1: Principles of PGNAA (On-line using prompt gamma) and NAA (Off-line using delayed gamma) Techniques

The activity or number of detected gamma rays of a particular energy is directly proportional to the disintegrations rate of the radionuclide, which in turn is directly proportional to the amount of parent isotope (target nucleus) in the sample. Thus, measurement of the gamma rays provides a measure of the total concentration of the parent element.

1.3 Radioactivity formed in NAA

The radioactivity formed (A) for an element X during neutron irradiation is given by basic equation for NAA, as follows

$$A = N\sigma\phi(1 - e^{-\lambda t_i}) \quad (1)$$

where A is activity in disintegration per second (dps), N is number of parent atoms, σ is neutron cross section, Φ is the neutron flux, λ is decay constant and t_i is irradiation time,

The activity after a decay period of t_d is given by,

$$A = N\sigma\phi (1 - e^{-\lambda t_i}) e^{-\lambda t_d} \quad (2)$$

where $S = (1 - e^{-\lambda t_i})$ is the saturation factor, $D = (e^{-\lambda t_d})$ is the decay factor,

The expression for the count rate (cps) is given by basic equation of NAA.

$$\text{cps} = (N_A \theta m / M) \sigma \phi S D C \epsilon \gamma_a \quad (3)$$

where, N_A is Avogadro's number, m is mass of the element, M is atomic mass of the element, ε is the absolute efficiency of detector for the particular gamma ray measured, γ_a is gamma-ray abundance and θ is isotopic abundance of parent isotope, $C = ((1 - e^{-\lambda CL}) / \lambda LT)$ is the counting correction factor, which takes care the decay parent during counting, CL is the clock time, LT is live time. When all the parameters determining the induced activity are known, the mass of the element (m) can be calculated, in principle, from the measured radioactivity. Greater the value of activity, the better will be the detection sensitivity for a given element. The nuclear cross sections for thermal neutrons via the (n, γ) process are generally higher relative to fast neutrons or other types of nuclear reactions. For this reason, thermal NAA is the sensitive method for most of the elements.

2. NAA Methods for concentration calculation

The following three methods of NAA are used for concentration calculations.

- Absolute method
- Relative method
- Single comparator or k_0 -based method

2.1 Absolute Method (Parametric)

The method of NAA which determines the elemental concentration in the sample directly from the activity produced is known as absolute method or parametric method. However, in absolute method, it is difficult to evaluate the exact values of σ and ϕ due their variation with energy of neutrons; therefore, this method is not used.

2.2 Relative method

This method is simple to arrive at the results, since it does not need the nuclear and reactor based input parameters as in the case of absolute method. In this method, a comparator / standard is co-irradiated with the sample and the activities from both sample and standard are measured in identical geometry with respect to the detector. From the activity ratios (cps) corrected by decay factors (D), the concentration of an element (m_x) is arrived as per the equation 4.

$$m_{x,sample} = m_{x,std} \times \frac{cps_{x,sam}}{cps_{x,std}} \times \frac{D_{std}}{D_{sample}} \quad (4)$$

Though the relative method is simple and precise, prior knowledge of the elements present in the sample is necessary to prepare corresponding standards in a similar matrix. In such cases, a method like single comparator or monostandard method can be used, which does not need a priori knowledge of the analytes in the sample.

2.3 Single Comparator or k_0 -based method

This method is based on the co-irradiation of the sample with a single comparator like Au, and the use of an experimentally determined composite nuclear constant k_0 [8]. The analysis results are linked to the k_0 factors, absolute detection efficiency and neutron spectrum characteristics. The concentration of an element is calculated using the following equation.

$$C(\mu g \cdot g^{-1}) = \frac{N_p / LT}{S.D.C.W} \cdot \frac{1}{\left(\frac{N_p / LT}{S.D.C.w} \right)^*} \cdot \frac{1}{k_0} \cdot \frac{f + Q_0(\alpha)^*}{f + Q_0(\alpha)} \cdot \frac{\varepsilon^*}{\varepsilon} \quad (5)$$

where N_p is the net counts, W and w are masses of sample and single comparator respectively, f is the sub cadmium to epithermal flux ratio, α is the epithermal neutron flux shape parameter, $Q_0(\alpha) (=I_0(\alpha)/\sigma_0)$ is the α corrected Q_0 and the k_0 is a factor and is taken from the literature [8]. The reactor-based parameters (f and α) are pre-determined for the irradiation position used for standardization purposes [7]. The symbol “*” refers to the comparator element.

2.4 The k_0 -based internal monostandard NAA (IM-NAA) method

The Internal Monostandard NAA (IM-NAA) is promising for analyzing large and non-standard geometry samples including small size samples [9,10]. This method utilizes an element present in the sample as monostandard and *in situ* relative efficiency by using gamma lines of activation products produced in each sample. The *in-situ* detection efficiency takes care of attenuation and geometrical effects of sample, and thus makes the method geometry independent. This approach gives elemental concentration of an element (x) relative to mono standard (y) as given below,

$$\frac{m_x}{m_y} = \frac{((S.D.C).(f + Q_0(\alpha)))_y}{((S.D.C).(f + Q_0(\alpha)))_x} \cdot \frac{P_{A,x}}{P_{A,y}} \cdot \frac{\varepsilon_{\gamma,y}}{\varepsilon_{\gamma,x}} \cdot \frac{1}{k_0} \quad (6)$$

where $k_0 = k_{0,Au}(x) / k_{0,Au}(y)$ and other terms are already defined. The relative concentrations are converted to absolute value by using monostandard mass obtained by any other method or NAA using a sub-sample analysis. In special cases like metals and alloys, where all the major and minor elements are amenable to NAA, absolute concentrations can be arrived using mass balance.

2.5 Prompt Gamma NAA (PGNAA)

When a thermal neutron is captured by the target nuclide, it is formed with excitation energy of about 2-8 MeV, depending on the binding energy of neutron. During the process of de-excitation of these nuclei, gamma-rays characteristic of the nuclide are emitted and are called prompt gamma rays. These gamma-rays are used for qualitative and quantitative assay of the target nuclei and technique based on this is called Prompt Gamma ray Neutron Activation Analysis (PGNAA). The PGNAA is a promising technique for the concentration determination of elements in varieties of samples [11,12]. The PGNAA method is complementary to conventional NAA and is suitable for most of the elements including low Z elements such as H, B, Si, P, S, Cl and Ti. The prompt gamma ray method is particularly useful for elements where product nucleus during normal NAA may have any of these difficulties, like (i) too short a half-life, (ii) too long a half-life, (iii) a stable nuclide like $^{12}\text{C}(n,\gamma)^{13}\text{C}$ and $^{10}\text{B}(n,\gamma)^{11}\text{B}$, (iv) a target nuclide having low isotopic abundance and (v) no delayed gamma ray from activation product like ^{32}P and ^{35}S . Non-destructive analysis of samples can easily be done by PGNAA, as the residual activity formed by irradiation of sample is less (due to low neutron flux about $10^{5-7} \text{ cm}^{-2} \text{ s}^{-1}$) and total counts depends on duration of assay / counting during irradiation. High neutron absorption cross section elements like B, Cd and Gd and major elements of samples of various matrices including alloys can easily be done by PGNAA.

3. Facilities for NAA

3.1 Neutron Sources

The neutron sources are primarily of three types; isotopic sources (^{252}Cf , $^{241}\text{Am-Be}$ etc), accelerator based (D-D or D-T) and research reactors. Some are sophisticated and extremely expensive, while others are rather simple and modest in cost. The most useful irradiation

facilities are provided by nuclear reactors where highest neutron yields (neutron flux) in the range of 10^{11} - 10^{15} n cm⁻²s⁻¹ are available as activation sources.

Thermal Neutrons: The nuclear cross-section for thermal neutrons in the (n, γ) process are generally high compared with the reaction capture cross-sections for neutrons of high energies, thus thermal NAA is an extremely sensitive method for most elements. This method has higher sensitivity for many elements except lighter elements such as C, N and O, and other elements such as Tl, Pb and Bi. Sensitivity varies depending upon (n, γ) capture cross section (σ) and half-life of radioisotope of interest, and many elements can be determined. This work is done using reactor neutrons having higher thermal neutron component or neutrons from graphite moderated thermal columns having thermal neutron component more than 99%.

Epithermal Neutrons: Activation Analysis using epithermal and resonance neutrons is known as Epithermal Neutron Activation Analysis (ENAA). Though epithermal neutrons have energies in the range of 0.2 to 1 eV, epithermal or more accurately epi-cadmium energy region (energy > 0.55eV, Cd cut-off energy) neutrons no longer follow the $1/v$ law closely. ENAA is highly sensitive and selective for those elements which have large resonance integrals (I_0). Elements with high thermal neutron cross sections are less sensitive towards ENAA and hence interference due to such elements get suppressed in ENAA, which will result in an improved detection limit and precision. Samples are covered with either Cd or B₄C for irradiation, though there are limitations in Cd and B amount for irradiation in a position of reactor. The Cd or B absorbs thermal neutrons and filters out epithermal energy neutrons. ENAA is mostly used for geological, biological or glass samples to suppress the high thermal neutron induced activities from matrix elements like Na. Elements like Ag, As, Au, Ba, Br, Cs, Ga, In, Mo, Pd, Pt, Rb, Sb, Se, Sm, Ta, Tb, W, Th and U are preferred for ENAA as these have higher resonance integral to 2200 ms^{-1} cross section ratios ($I_0/\sigma_{\text{th}} = Q_0$; $Q_0 > 10$).

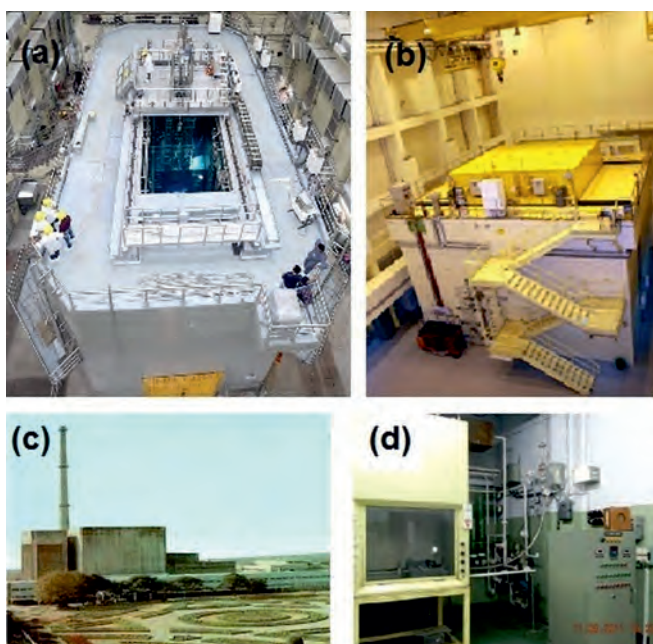


Figure 2: Facilities for neutron irradiations (a) Apsara-U reactor, (b) Critical facility (CF) reactor, (c) Dhruva reactor, (d) Pneumatic carrier facility (PCF) at Dhruva reactor, BARC

Fast Neutrons: When the measurement of elements especially light elements such as C, N and O) and other elements is not favorable by thermal neutrons because of the very low (n,γ) cross-sections, the fast neutrons can be used for determination elements using prompt or delayed gamma-rays produced from (n,n') , $(n,2n)$, (n,α) and (n,p) type nuclear reactions [4]. Apart from reactor, Isotopic neutron sources (like $^{241}\text{Am-Be}$) or Cockcroft-Walton type accelerator using the reaction D-D and D-T reactions can generate fast neutrons 2.45 and 14 MeV, respectively.



The advantages of Fast NAA are that; it is fast and non-destructive, can be used on a routine basis, is ideal for short irradiations, and is particularly sensitive for detection of light elements. The disadvantage of this method is it requires considerable shielding because of high-energy neutrons.

Neutron irradiation facilities currently available at BARC, Trombay include Dhruva reactor, Apsara-U reactor, Critical facility reactor [13] and D-T neutron generator at Purnima. Some of the neutron irradiation facilities utilized for NAA work are given in Fig. 2. The Pneumatic Carrier Facility (PCF) of Dhruva reactor is utilized during neutron irradiation of samples for R&D work and routine sample analysis using NAA [14]. PCF is useful for those elements namely Al, V, Ti, Mg, Ca, Mn, Na, K and a few REEs whose activation products are short and medium lived (half-lives in the ranges of minutes to hours). The turnaround time of analysis is less as it involves short irradiation and faster counting of irradiated samples. The available neutron fluxes from thermal column to core irradiation positions of research reactors at BARC are in the range of 10^7 - 10^{14} $\text{n cm}^{-2}\text{s}^{-1}$.

3.2 Gamma-ray Detectors

Commonly used detectors for gamma ray measurements are high purity germanium (HPGe) detector as well as NaI (TI) and Bismuth Germanate (BGO) detectors. HPGe detector is widely used during conventional NAA (thermal neutron activation) and PGNAA due to better resolution. For Compton suppressed detection system HPGe-BGO or HPGe-Nai(Tl) is suitable in anti-coincidence mode. Lanthanum Bromide (LaBr_3) is also used for low as well as high energy gamma-ray spectrometry, as it has better resolution compared to BGO. Typical resolutions of HPGe detector is about 0.12% at 1332 keV gamma ray of ^{60}Co . NaI(Tl), BGO and LaBr_3 detectors can be operated at room temperature making them suitable for field applications and their resolutions at 662 keV gamma ray of ^{137}Cs are about 10%, 12% and 4%, respectively.

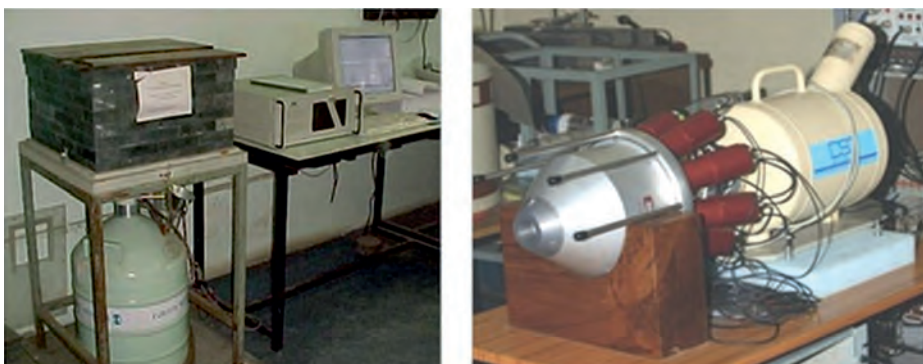


Figure 3: Typical gamma-ray spectrometry setup (a) singles counting using HPGe detector coupled to MCA and (b) Compton Suppressed system using HPGe-BGO detectors

4. Experimental Methodologies

Steps involved in NAA include, sampling and sample preparation, sample irradiation in suitable neutron source, measurement of gamma radiations from the activation products, peak area calculation and concentration determination. Both solid and liquid samples can be analyzed using NAA technique. Solid samples are either packed in polyethylene sheet or aluminum foils for irradiation depending on the neutron flux and duration of irradiation. When comparator method of NAA is used, samples and standards are packed in similar way and co-irradiated in neutron source such as nuclear reactor. Many samples and standards can be irradiated together followed by measurement of gamma rays from each sample separately in identical geometry in the detector. Peak area analysis can be done using suitable peak fit software. Concentrations are arrived using either relative method or k_0 -method as mentioned earlier. Both small sized samples (a few tens milligrams to less than a gram) to large and nonstandard geometry samples can be analyzed.

4.1 Methodologies in NAA

Methodologies of NAA involves direct irradiation of solid samples without any destruction or sample dissolution by INAA or it can be performed with pre or post irradiation chemical separations by PNAA (also called as chemical neutron activation analysis, CNAA) and RNAA, respectively. INAA is a nondestructive method where as CNAA and RNAA are destructive methods. In INAA the sample is irradiated and counted after giving proper cooling time. This method is nondestructive as well as multi elemental. This method cannot be used if there is spectral interference, if indicator isotope is pure beta emitter and if the background is too high (which masks the analyte peak). In CNAA the analyte is pre-concentrated before irradiation. The advantage here is that higher amounts of the sample can be used for preconcentration. This method is particularly useful for testing high purity materials (>99.99 %) i.e. to test for impurities in pure material, preconcentration is advantageous. In some cases, the radionuclides from the (n, γ) process also produced from other nuclear reactions like (n, f) , e.g. during determination of lanthanum in uranium matrix, desired radionuclide is produced by the reaction $^{139}\text{La}(n, \gamma)^{140}\text{La}$. ^{140}La and other light REE are also produced due to fission of uranium. Therefore, the results become erroneous. In such cases also CNAA is useful i.e. separation of lanthanum and other REE from uranium matrix before irradiation. CNAA methodologies were developed for impurities in high purity Ga/As [15], Sc [16] and speciation of arsenic in ground water [17]. In RNAA the separation of matrix is done after irradiation. This method is preferred over CNAA when blank interference has to be avoided (once irradiated, sample becomes radioactive while the reagents used for separation are inactive). This method is utilized during analysis of forensic exhibits.

4.2 Quality Assurance/Quality Control (QA/QC)

Certified/Standard Reference Materials (CRMs/SRMs) are used as 'control' sample for quality control (QC) purposes to validate analytical methods by evaluating the accuracy of determined value with respect to the certified/recommended values by the certifying agencies. In the cases of non-availability of CRMs, results can be checked by other reference analytical techniques. Quality Assurance (QA) involves, QC along with other parameters like precision or reproducibility of data from replicate sample analysis, total uncertainty measurements and detection limits of elements by the method used. For validation of the relative, k_0 -based NAA and IM-NAA, various RMs/CRMs/SRMs having matrices like geological, biological, soil, sediment, steel, stainless steel and Ni-alloy from IAEA, NIST, USGS, BCS and IRMM as well as synthetic multi-element standards (SMELS) were analyzed [10, 18, 19]. The accuracy of the

method was evaluated from percentage deviation and Z-score values. Percentage deviations from certified/recommended values were in the range of ± 0.5 -10% and the Z-score values for many elements determined were within ± 1 at 95% confidence level.

4.3 Sensitivity & Detection Limit in NAA

Detection limits by NAA methods are calculated using sample background and elemental sensitivity. Depending on background counts and elemental sensitivity (cps/ μg), the detection limits vary from ppb (ng/g or $\mu\text{g kg}^{-1}$) to ppm ($\mu\text{g/g}$ or mg kg^{-1}) [1, 4]. For example Dy, Sm, Eu, Mn, Au and Sc can be determined at ppb concentration level by NAA whereas H, B, Cd, Gd and Dy can be determined at ppm level by PGNAA using thermal neutron beams from reactors and C, N, and O can be determined at major concentrations (percentage level) by Fast NAA using 14 MeV neutrons. Details of experimental detection limits obtained for various samples can be found in referred literature in this article.

5. Applications of NAA

NAA technique has been extensively applied for chemical characterization of wide varieties of samples related to geological, environmental, biological, forensic and archeological origin as well as materials sciences [13, 14, 20-57]. A few of the typical applications from our R&D work are summarized in the present article.

5.1 Geological samples, ores/minerals and gemstones

Neutron activation analysis is a suitable technique for major, minor and trace constituents of geological samples including various ores, minerals during exploration studies. Uranium ores [22], zircon, and rutile minerals were analyzed for uranium concentrations (200-500 mg kg^{-1}) by using small as well as large sample NAA. Geological rocks and meteorites have been analyzed for U, rare earth elements (REEs) and precious element concentration determination by NAA [23].

Elemental compositions of a meteorite sample: Meteorites belong to rare and precious materials, and are useful for studying origin and evolution of our planetary system. One of the main criteria for identification and classification of meteorites is their chemical composition. A new meteorite sample named as Jagannath meteorite, collected from the impact on 27th September 2003 in Odisha state, India, was analyzed by the IM-NAA method [24]. Concentration values of 20 elements including major, minor and trace elements were determined. The meteorite was found highly enriched in elements like Mn, Cr, Co, and Ni including Fe compared to the elemental abundances of the earth crust. Elemental abundances of some elements present in the analyzed meteorite compare well with chondrite values and with two recent meteorites namely the Kobe meteorites and the Czech Moravka meteorites. The data on chemical composition compared well with these two meteorites and confirmed that the present sample analyzed is an authentic meteorite.

Analysis of emeralds and associated rocks: Natural emeralds and their associated rocks along with beryl samples were analyzed to investigate the process of formation of emeralds. The k_0 -based NAA method was utilized for the quantification of 21 elements [25]. From the data the following observations were made: (i) The aluminum content, determined using NAA, in emerald and beryl were 10.02 % and 10.03 %, respectively which agreed well with the expected concentration of aluminum in emerald and beryl (~ 10 %), beryllium aluminum silicate [$\text{Be}_3\text{Al}_2(\text{SiO}_3)_6$], (ii) Concentrations of Cr, Cs, Sc, Rb and V were higher in emerald compared to the associated rocks, (iii) Our studies showed the presence of magnesium as a major and cobalt as a trace constituent in emerald, (iv) the chromium content of the emerald is the highest and it

decreases in the order, emerald > trapped rock > host rock, indicating migration of Cr from associated rock to emerald and (v) Potassium was found to be absent and Na was present in varying concentrations in emerald and associated rocks.

Analysis of Beryl samples: Beryl samples, collected from Geological Survey of India (GSI), Pune were analyzed for trace elements. Concentrations of the analytes in samples are shown in Fig. 4. Concentrations of Mn, Fe, Cr and rare earths were found to be higher in red beryl compared to colorless beryl, pale green (PG) and yellowish green (YG) beryl samples. INAA method found to be useful for distinguishing the red beryl samples from all other beryl samples by just analyzing the elemental profiling.

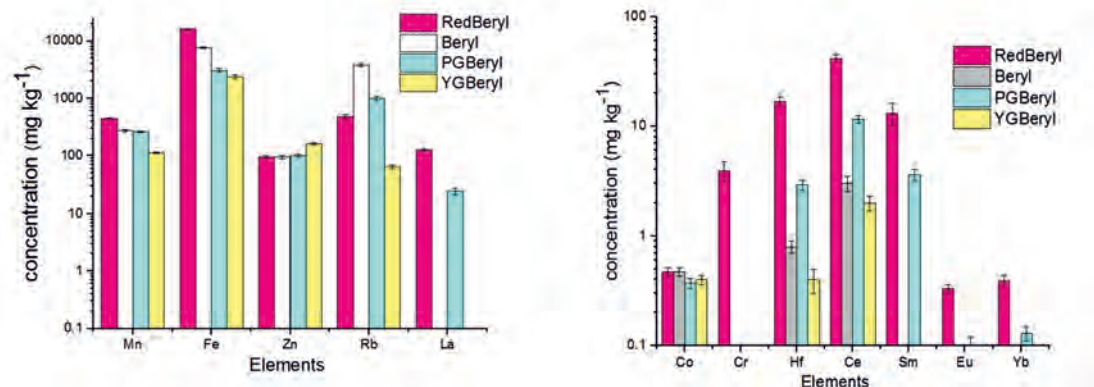


Figure 4: Concentrations of elements in Beryl samples

Analysis of precious stones (gemstones): Natural ruby can be discriminated from the synthetic or artificial ruby, as it contains many multi-elements from major to trace concentration level. The k_0 -NAA was applied for determination of concentrations in two natural and one synthetic ruby samples [7]. The trace element profile was used to characterize the ruby samples. A total of 22 elements were determined in two natural rubies, whereas only 7 elements in synthetic ruby confirmed that the two ruby samples are of natural origin. INAA is a potential technique for certifying gemstones (like ruby) and diamonds from various origins whether they are of natural or artificial/synthetic origin.

Analysis of uranium ores: Small and large size samples of uranium (U) ores were analyzed using the IM-NAA method [22]. Correction factors due to fission products of U like La, Ce, Nd, Zr and Mo to avoid spectral interferences, were obtained using elemental standards. Accuracy of the IM-NAA method was assessed by analyzing the coal fly ash NIST SRM (1633a) with low U content. For this purpose, NIST SRM 1633a was spiked with U (~0.05%) and analyzed. The method was used for the analysis of uranium ore samples for multielement contents including U and Th.

5.2 Environmental Samples for total elemental concentration and trace element speciation

Environmental sample analysis is a key tool in the effective management of our environmental resources and human health conditions. NAA technique is routinely used for analysis of various environmental matrices like soils, sediments, coal fly ash and tailings of various industrial samples for many elements. NAA was used for multielement determinations for studying mobilization of elements in water and sediments of Satluj River and Harike Wetland, Punjab region, India [26] as well as for quantifying toxic and rare earth elements in

sediment samples from Nainital Lakes [27] and Periyar Lake [28]. The knowledge of sedimentation rates and the elemental concentration levels in sediments from lakes and water bodies can provide clues for the pollutants. The concentrations elements were determined by k_0 -NAA method in this dated sediment [27]. Elements like Cr, Fe, K, Mn and Zn are found to be in negative correlation with depth (1-51 cm), whereas elements like As, Ba, Cs and Na indicated positive correlation.

Speciation of As and Cr in water samples: Studies on arsenic is important due to its toxicity. Ground water samples were analyzed for total As and inorganic species i.e. As(III & V) by INAA [29]. Anion exchange separation methods were standardized for the speciation of As(III) and As(V) using radiotracers with Dowex 1X8 resin in chloride as well as acetate forms. The method was validated using synthetic mixtures of arsenic species, As(III) and As(V), and applied to water samples. Arsenic speciation of water samples revealed that As(III) is about twice as high as As(V) concentrations. Membrane based separations in conjunction with INAA have shown promising results for quantifying low concentration levels of species of As (V) and Cr (VI). The detection limit of 3 ppb was achieved by membrane based NAA is well below the limit (10 ppb) set by WHO in drinking water.

5.3 Application to Biological, Food and Agricultural Samples

Various biological materials such as leaves, blood, ayurvedic formulations [30] were analyzed using NAA technique. Environmental exposure to human was studied by sampling hair from Indian student population and analyzing the trace elements using NAA [31-32]. Mercury concentrations were evaluated in Indian prawn using NAA for export purposes [33]. Elemental separation procedure was standardized for quantification of As, Mn, Mo, Cu and Zn milk sample using NAA [34]. Wheat grass [35, 36], herbs, plant materials, muscles/bones were analyzed for trace elements using NAA. Renal calculi from patients of Stone belt region, India were analyzed for various elements present in trace and ultra-trace levels using NAA technique [37] and studied for possible correlation of diet and renal stone formation. The phyto-accumulation efficacy of chickpea plant for selenium (Se) was assessed by determining Se concentration in soils and various parts of chickpea plants grown in those soils [38]. NAA technique has been extensively utilized for essential / toxic element determination in wide varieties of food samples including rice, wheat, fish. NAA methodology has been standardized for the determination of concentration of iodine in grass and cow milk to study the transfer of iodine from grass to milk [39].



Figure 5: Wheat grains utilized for the elemental quantification through INAA

Application to large and small size wheat samples: The IM-NAA methodology was used to analyze several small and large size samples of wheat grains [40] in order to examine the level of in-homogeneity in small size sample and also to obtain a representative sample mass to get accurate results. The results of small size samples (50 mg to 1 g) showed large variations (> 22%), whereas the relative concentration values of three larger size samples (0.45-1 kg) indicated a good agreement (%RSD 0.3-10%). These results suggest that analysis of one large sample is sufficient instead of replicate sub samples. Varying amount of sub sample analysis indicated that a sample mass of 1 g and above is adequate or representative for analysis.

Application of Epithermal NAA (ENAA) for quantification of Iodine food samples: Quantification of iodine (using ^{128}I , 25 min, with higher resonance integral, $Q_0 \sim 25$) is difficult in the presence of interfering elements like Cl, Mn, K and Na. To suppress the thermal activation of interfering elements, ENAA method was standardized using a B_4C filter for total iodine determination at trace concentration levels in various food and food products [41, 42]. The concentration of iodine could be determined even in iodized packaged salts from market. The results showed that iodine can be determined at trace concentration levels in the presence of high salt content without chemical separations. Iodine concentrations were also measured in different biological reference materials obtained from IAEA and NIST for quality control (QC) purposes. The detection limits of iodine for various food-stuffs were in the range of 0.02-0.6 mg kg^{-1} whereas the detection limit for iodized salt sample was $\sim 5 \text{ mg kg}^{-1}$.

Application of INAA, chemical NAA for total selenium and its bioaccessibility in food and agricultural samples: INAA has been successfully applied for quantification total Se in seleniferous soil samples from Punjab [43]. It was of interest to study total Se and Se(VI) concentrations in food grains (wheat), mustard and alliumcepa (onion) by Instrumental or chemical NAA [44, 45]. Detailed studies were carried out in which NAA has played a major role.

5.4 Archaeological objects analysis

Compositional analysis in archaeology involves the analysis and interpretation of chemical fingerprints obtained from archaeological materials. Chemical compositions analysis is an important tool for provenance study of archaeological artifacts like pottery, brick, glass and ceramics. INAA was utilized for the analysis of different archaeological artifacts [46-48]. Under an IAEA Coordinated Research Project (CRP) on large sample NAA (LSNAA), an irregular shape and larger size clay pottery was analyzed, in which a total of 18 elements were determined with respect to Na as internal monostandard [47].

Application of INAA for provenance study of ancient pottery samples: Several pottery samples collected from different excavated Buddhist sites of India [46] were analyzed by IM-NAA. Concentrations of 20 elements consisting of Al, alkali, alkaline and transition



Figure 6: Typical clay ceramic artifacts (pottery and brick) from Buddhist sites of India analyzed by IM-NAA for provenance study

elements were determined in pottery as well as soil samples from the same location. Elemental concentration ratios of Al to Sc and concentrations of some key elements namely Na, K, Rb, Cs and Ba as well as Sc, Fe, Co, Cr, Hf and Mn were used to establish the provenance/grouping of pottery artifacts, i.e., whether they belong to same of different sources/origin. REEs in these pottery samples were determined and their concentrations were used for grouping. Provenance / grouping studies could be carried out by a standard-less approach using elemental concentration ratios with respect to Sc (normalizing element) using IM-NAA, instead of absolute concentrations.

5.5 Forensic Applications

In general, forensic science is the utilization of science to solve the criminal ramifications by analyzing the physical evidences. There are many analytical techniques that are being practised in various forensic laboratories. Beside all conventional analytical techniques, nuclear analytical technique such as NAA [49] has been used for analysis of forensic evidences, which mainly include the analysis of hair, nails, viscera samples, narcotic samples, gunshot residues (GSR) [50], transmission wires, paint, food samples and glass or ceramic samples and trafficking of high purity/precious materials such as ornaments. The marker elements for GSR are Sb, Ba and Cu and quantified by adopting radiochemical approach. Bullet pieces were analyzed for trace elements for source correspondence cases. Viscera samples from suspected poisoning cases were analyzed for marker element zinc using NAA [51]. Various transmission wire (Cu / Al) were analyzed for trace element profiles for source correspondence. Ag, Au, Se, Sb in aluminum and Mn, Cr, Hf, Sc, Fe, REEs in copper wires are indicative for such investigations. NAA is also used for the food and glass forensics. Food forensic specify the certification of authentic food products by determination of essential or toxic elements in various commonly used food products like baby food, commercial packed food stuff, meat and fish. Minor and trace elements were estimated in raw and branded turmeric samples using INAA [52]. Fig. 7 gives a gamma ray spectrum of neutron irradiated turmeric sample. Concentrations of the most the elements were found to be within the safe limit set by WHO including Cr (2.3 mg kg⁻¹).

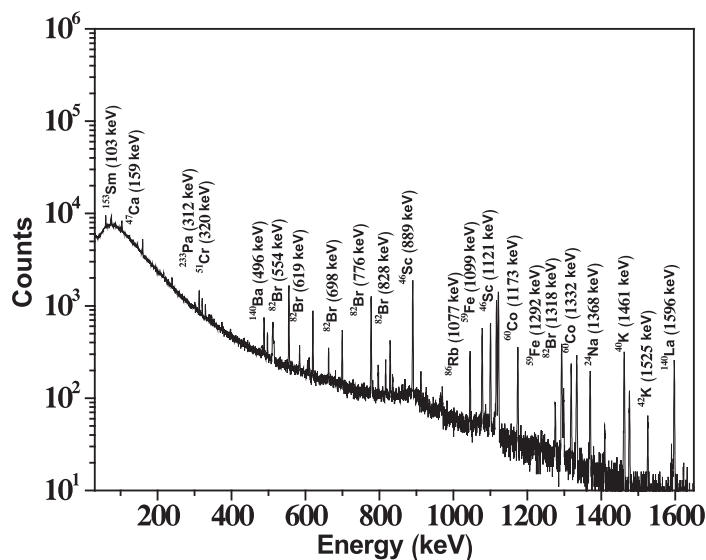


Figure 7: Gamma ray spectrum of neutron irradiated raw turmeric sample

Glass Forensics: Glass fragments, related to container glass, automobile/windshield glass or window pane glass, are potential forensic evidences in various types of anti-social activities and accidents. INAA is an important technique to get the information on elements of interests from major to trace concentration level. Automobile (Sodalime) glass is mainly composed of elements Si, Na, Ca, Mg and Al at percentage level and transition and rare earth impurities are at trace concentration level. Elements at minor and trace concentration levels are regarded as the key / fingerprinting elements for various glass samples. INAA was utilized for the quantification of fourteen elements including ten trace elements [53, 54]. Elemental concentration ratios like La/Ce or La/Sc or total REE are useful for preliminary grouping the similar class of glasses obtained from same or different brands of automobile (e.g. car). The results of the minor and trace elements (Mn, Sc, Co, Zn, Zr, Ta, Hf, La, Ce, Sm and Eu) present in five different automobile glass samples from three different brands could be utilized for grouping the glass samples. A typical grouping pattern obtained by statistical cluster analysis using minor and trace elements is shown in Fig. 8. It indicated that the five automobile glass samples are broadly grouped into three groups; Group 1 (Glass 1), Group 2 (Glass 4 & 5) and Group 3 (Glass 2 & 3).

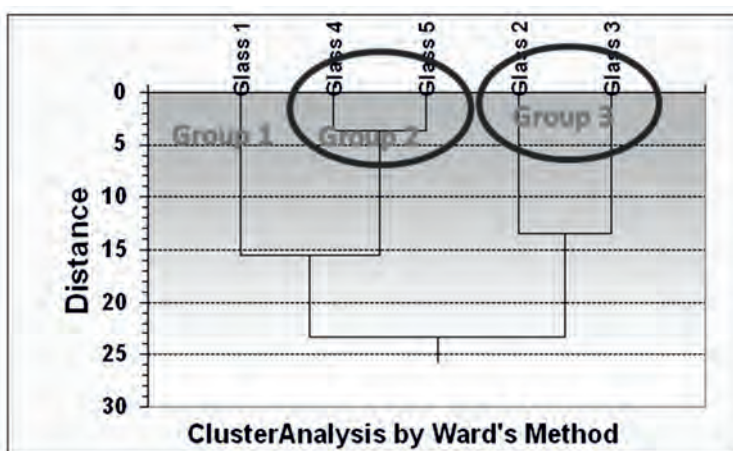


Figure 8: Tree dendrogram using minor and trace elements employing Ward's method

5.6 Application Materials Sciences and high purity materials

Compositional characterization and trace element determination in materials developed or procured or synthesized is very important under chemical quality control (CQC) exercise for their acceptance in the intended use. Compositional characterization was carried out in various alloys having nuclear and non-nuclear applications. NAA methods were developed and utilized for the determination of elements (impurities) in high pure Al, Ga and As [15] and Sc [16] as well as in Si/SiO₂. NAA was used for the determination of tungsten in steel and chlorine concentration at trace levels in zirconium niobium alloys.

Analysis of Ta-W Alloy: Tantalum-Tungsten alloy, is used for various purposes like corrosion resistant chemical processing equipments, heat exchangers and condensers. Evaluation of its chemical composition is important for suitable applications. Composition of Ta-W alloy was evaluated using relative method of NAA as well as IM-NAA (using mass balance) [13].

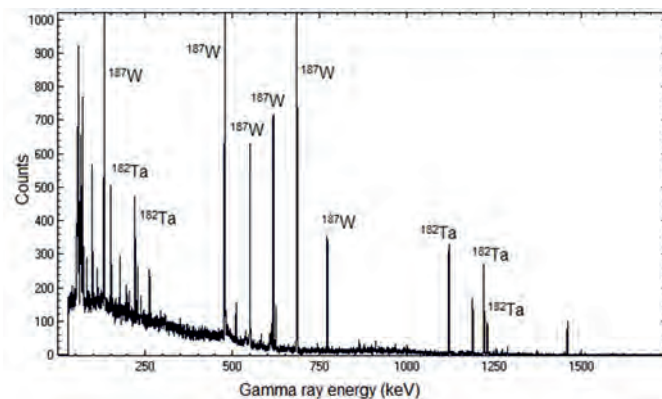


Figure 9: Gamma-ray spectrum of a neutron irradiated Ta-W alloy

Application to zircalloys and stainless steels: The IM-INAA in conjunction with in situ detection efficiency is useful for the analysis of metals and alloys of irregular size samples (Fig. 10) like zircaloy-2, zircaloy-4, and stainless steel (SS-316M) [9, 40, 55, 56]. Samples were irradiated in the thermal column (for large size) as well as in the core position (for small size) of the Apsara reactor. The corresponding neutron fluxes are about $2 \times 10^8 \text{ cm}^{-2} \text{ s}^{-1}$ and $5 \times 10^{11} \text{ cm}^{-2} \text{ s}^{-1}$



Zircaloy-2



SS-316M

Figure 10: Large and irregular shaped alloy samples analyzed by IM-NAA

Table 1: Determined concentrations of elements in zircaloy-2 and stainless steel (SS-316M) samples by IM-NAA

Zircaloy-2			SS-316 M		
Element	Concentration	Specifications	Element	Concentration	Specifications
Zr (%)	98 ± 1	Balance	Fe (%)	65 ± 1	Balance
Sn (%)	1.60 ± 0.03	1.2-1.7	Cr (%)	17.1 ± 0.1	16.5-17.5
Fe (%)	0.18 ± 0.02	0.07-0.2	Ni (%)	13.5 ± 1.3	13.00-14.00
Cr (%)	0.085 ± 0.001	0.05-0.15	Mo (%)	2.3 ± 0.1	2.00-2.75
Ni (%)	0.075 ± 0.003	0.03-0.08	Mn (%)	1.88 ± 0.01	1.5-2.0
Co*	7.0 ± 0.1	20	Co*	145 ± 4	300 max
Ta*	16 ± 1	200	As*	53 ± 3	NA
Hf*	25.1 ± 0.8	100	W*	26 ± 3	NA
Mn*	11.0 ± 0.1	50			
As*	5.1 ± 0.4	NA			

* concentration in ppm (mg kg^{-1}); NA – not available

respectively. The results of a zircaloy-2 and stainless steel (SS-316M) are given in Table 1. Zr and Fe present in the samples were used as internal monostandard in zircalloys and stainless steels, respectively. In addition to major and minor elements in zircalloys and stainless steels, some trace elements like As, Co, Sn, Ta and W were quantified. In the presence of major element of Zr, trace Hf could be determined in zircaloy-2 sample. Since all the major and minor elements in zircalloys and stainless steels are measurable by NAA, concentrations were obtained by a standard-less approach (mass balance procedure) by using IM-NAA method [9].

5.7 Industrial Applications

Analysis of coal: Coal and cement quality is important for industrial applications. Coal analysis generally deals with the estimation of C, O, H, S and Ash content (oxide of Si, Al, Fe, Ca, Ti, Mg and Na). Beside this, it also includes the quantification of major constituents such as C, H, O, N, S; that ultimately classify the coal on the basis of its quality. Various techniques like ED-XRF are being utilized for ash content of coal. The light elements (C, H, N, and O) can also be determined using fast neutron activation techniques and marker elements of ash content in coal were analyzed using thermal neutron activation followed by prompt gamma ray measurements. IM-NAA was used for analysis of larger size sample of coal [22]. Trace and toxic constituents in coal were also determined using conventional NAA. Fast NAA using neutron generator (14 MeV neutron) is utilized for C, N and O determination whereas thermal neutron from this source are helpful for H, S, and coal ash elements (Si, Al, Fe and Ca). Since a long, isotopic neutron sources like ^{252}Cf and $^{241}\text{Am-Be}$ are used for coal ash contents determination by on-line NAA (PGNAA) using scintillation detectors like NaI(Tl) or BGO.

Ceramic catalysts: Platinum group metal catalysts are widely used in chemical process. Cordierite (magnesium aluminum silicate) supported platinum/palladium catalyst is a promising catalyst for hydrogen mitigation during loss-of-coolant accident conditions in nuclear reactors. The quality and specified elemental concentrations are important and thus their accurate determination is a necessity. The materials are highly refractory in nature. As INAA does not require sample dissolution, various catalyst samples (Fig. 11) were analyzed for platinum and palladium using INAA technique [13].

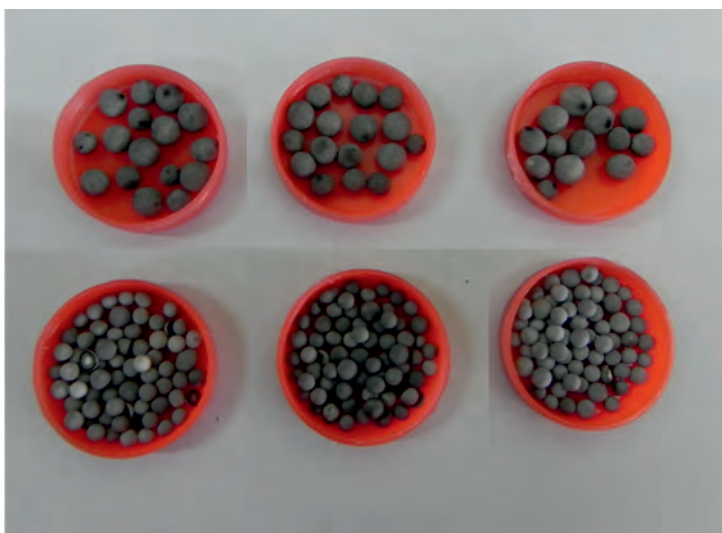


Figure 11: Catalyst samples analyzed for Pt/Pd

Precious metals in waste materials: Fire clay crucibles are used for refining of gold (Au) and other precious metals and these metals diffuse into the walls of crucible during refining process. The crucibles are crushed at the end of the process into pieces and the ground material is termed as dross. As the dross contains substantial amount of metals like Au and Ag, thus from precious metal recovery as well as accounting point of view, quantification of Au and Ag in dross is required. Dross is usually heterogeneous with respect to gold and silver. Instead of many replicate sample analysis, large size sample analysis was carried out by IM-NAA for quantification of gold (200-400 mg kg⁻¹) and silver (1200-1700 mg kg⁻¹) in different dross samples [57]. The larger size samples (about 100 g or more) were irradiated at low flux position of Critical Facility reactor.

5.8 Research and Development work on PGMAA

PGMAA facility was set up using thermal neutron beam facilities of SSPD at Dhruva reactor, BARC [58]. The method is useful for major/minor concentrations in solid samples and trace concentrations of neutron poisons like B, Cd and Gd. The k_0 -based PGMAA was standardized by evaluating prompt k_0 -factors of different isotopes H, B, Mg, Si, S, K, Cd, Sm, Hg and Gd with respect to ³⁶Cl (1951 keV prompt gamma-ray). The method was applied to reference materials [11] and samples like meteorites and alloys. Cement standard and samples were analyzed as a part of IAEA inter lab comparison exercise [59] and the results (Ca, Si, Fe, Al etc.) were found to be in good agreement with the recommended values. A method was developed for correcting neutron self-absorption using sample mixed with NH₄Cl [60]. The developed method was used to determine B in boron based chemical compounds [60, 61] and borosilicate glass samples. Elements like B, Ti, Mo, Cr and Si were determined in TiB₂ alloy. Total boron concentration was determined in boron-based composites and boron carbide samples utilizing thermal neutron beam at Dhruva and using 478 keV from ¹⁰B [61]. Boron was determined using 478 keV ¹⁰B(n,αγ)⁷Li. Hydrogen concentrations in Zircaloy samples can be determined utilizing 2223 keV prompt gamma ray from ¹H(n_{th},γ)²H reaction. Further details of PGMAA can be found from references 62 and 63.

6. Conclusions

Neutron Activation Analysis is an important nuclear analytical technique, which has been extensively utilized for materials characterization in many fields of Science and Technology due to its several advantageous properties. It fulfils almost all the requirements under QA/QC. Though, NAA has been used extensively in nuclear technology, the present article deals with various non-nuclear applications that are relevant for societal benefits. NAA finds enormous applications in various fields having societal applications like speciation, bioaccessibility of trace elements and multielement determination in samples of environment, geology, biology, food, agriculture, industry, archaeology and forensic sciences. INAA finds applications in high purity materials and material composition in alloys, glass and ceramics, that are difficult to dissolve and analyze by conventional analytical techniques. PGMAA is a truly non-destructive technique, which can give quantitative information on H, B, many other elements of interest in finished products and the sample can be returned after analyses, which are important in many cases such as archaeology, forensic and materials sciences. The NAA technique, in general, has got enough scope for R&D works having societal applications under various collaborative schemes with academic as well as research institutes.

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