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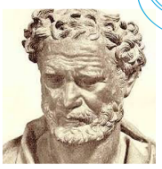
Hellenic Neutron Association Newsletter



Editorial

In the second newsletter of HENA the status and prospects of neutron activation analysis, a powerful technique for multi-elemental analysis, are presented by Dr Ion Stamatelatos, whereas Dr Kalliopi Kanaki elaborates on the new generation neutron detectors for the European Spallation Source currently under construction in Lund, Sweden. We hope you enjoy reading it. Have a nice summer holidays!

— Dr. Konstantina Mergia (NCSR Demokritos)

Neutron Activation Analysis: Status and Prospects

by DR. ION E. STAMATELATOS *

Neutron Activation Analysis (NAA) is a nuclear analytical technique enabling non-destructive multi-elemental analysis in a variety of sample matrices with excellent sensitivity, accuracy, and precision [1]. The technique is based on irradiation of the sample under a known neutron field and detection of the characteristic gamma rays that are emitted as a result of neutron induced nuclear reactions in the sample matrix. The elemental composition of the sample material is determined by analysis of the detected gamma rays. It has been reported that about 70% of the stable isotopes in the nuclide chart can be determined by NAA techniques. The different NAA techniques are summarized in Table 1.

The realization of NAA requires knowledge of the incident neutron energy and fluence, the occurring nuclear reactions and their cross-sections, as well. The neutron energy determines the type of nuclear reaction as well as its probability and therefore defines the radioactive product(s). NAA has been performed utilizing thermal, epithermal, and fast neutrons produced by all types of available neutron sources (nuclear reactors, accelerators, isotopic sources). Nevertheless, the optimum measurement sensitivity is achieved in nuclear research reactors due to the significantly higher neutron fluence rates in these facilities. To analyze the complex gamma ray spectra encountered, the utilization of gamma ray detectors with a high energy resolution is required and most commonly this is achieved by using high purity germanium semiconductor detectors. Absolute, comparative, internal mono-standard and k0 techniques have been developed, to relate the net peak areas in the gamma-ray spectrum to the amounts of the elements present in the sample under given experimental conditions. The NAA method has considerable advantages. First, it is non-destructive, allowing reusing of the sample. Moreover, since it requires minimum sample preparation, the probability of laboratory contamination is minimized. Furthermore, its linearity ranges up to 12 orders of magnitude. The broad range of linearity is a particular advantage, since it allows a wide range of elements and concentrations to be detected

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and therefore allows the determination of major, minor, and trace elements in the sample simultaneously. In some matrices the measurement can be delayed to allow interfering radionuclides of a shorter half-life than the radionuclide of interest to decay. Consequently, interferences can be eliminated and lower detection limits for the radionuclides of interest can be achieved. Depending on the element of interest and the material matrix, the sensitivity of the technique may be of the order of parts per billion (ppb), the accuracy in the region of 5 % and the precision is often better than 0.1 % [1].

Prompt Gamma Neutron Activation Analysis (PGNAA)	Sample irradiation and gamma measurement are performed simultaneously
Delayed Gamma Neutron Activation Analysis (DGNAA)	Gamma-ray measurement starts after the end of irradiation
Cyclic Neutron Activation Analysis (CNAA)	Periodic irradiation, transfer and measurement of the sample for an optimum number of cycles
Instrumental Neutron Activation Analysis (INAA)	No sample treatment is involved
Radiochemical Neutron Activation Analysis (RNAA)	The sample is subject to chemical separation in order to remove interfering radionuclides or to concentrate the radionuclide of interest
Large Sample Neutron Activation Analysis (LSNAA)	Non destructive, non-invasive analysis of whole samples with masses in the range of 10 g up to several Kg

Table 1. NNA techniques

NAA has found successful applications in many areas of research and technology [2]. For example, in archaeology NAA is used to analyze objects and archaeological findings for provenance and authenticity investigations [3]. In material science, NAA has been used for the determination of the elemental composition of metals and inorganic compounds. In food and nutrition, NAA has been applied in order to determine essential elements in plants, animals and fishery products. In environmental studies, NAA has been applied to determine elements in soil and sediment. Moreover, it was applied to characterize aerosol collected on air filters. In geology, NAA was used to analyze rocks and cosmic materials. In biomedicine it was used for the analysis of toxic and essential elements in human tissues. Of particular interest was the application of NAA for the in vivo analysis of the human body composition providing unique results that cannot be obtained by any other available technique [4]. Examples are the in vivo determination of Cd and Hg in critical organs of occupationally exposed persons or the in vivo determination of major human body compartments, such as protein and fat, for nutritional and metabolic studies. In conventional NAA the mass of analyzed samples is in the range of milligrams to grams (or of a few μl to ml in volume), representing only a small portion of the bulk material. However, there are applications where larger samples need to be analyzed. For example, in authenticity and provenance studies the analyzed objects are usually precious, non-replaceable artefacts that need to be preserved intact and cannot be damaged for sampling purposes. Moreover, in the case of highly heterogeneous materials, as for example marine sediments, representative sampling is usually a very difficult or even an impossible task. For representative analysis of such samples Large Sample Neutron Activation Analysis (LSNAA) was developed [5]. In LSNAA, the non-destructive, multi-element character of the conventional NAA technique is combined with the ability to analyze bulk objects, up to several liters in volume, as a whole. The large sample is irradiated in a neutron field (Fig. 2) and is subsequently transferred to a suitable gamma-ray spectrometry facility to be counted either as a whole or using segmented scanning (Fig. 3). The gamma-rays emitted from the activated material are identified and the activity induced in the sample volume is quantified. Finally, the spectra analysis results are interpreted in terms of elemental concentrations. The parameter that differentiates LSNAA from the INAA technique is the large volume of the sample. To account for the volume of the sample, corrections factors are derived for neutron-self shielding during sample irradiation, gamma-ray self attenuation during gamma-counting, volume distribution of activity within the large sample, irregular sample shape and material inhomogeneity effects.

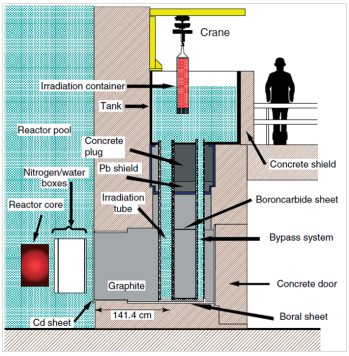


Fig. 2 LSNAA irradiation system at IRI reactor, TU Delft, The Netherlands

It is stressed that the analytical information obtained by LSNAA is representative of the bulk sample material and not the surface only. This is of particular importance, especially in cases of archaeological findings that may have been buried in the earth for a prolonged period resulting in erosion or distortion of their surface composition. Other techniques such as XRF or PIXE can only analyze superficial layers of the object and thus provide limited information on the

sample matrix composition in depth. In addition, LSNAA can be performed using facilities with much lower neutron fluence rates ($10^6 - 10^9 cm^{-2}s^{-1}$) than for 'conventional' NAA ($10^{12} - 10^{13} cm^{-2}s^{-1}$), since in LSNAA the 'loss' in the neutron flux is compensated by the greater mass of the analyzed sample material. Therefore small and medium power reactors, reactor thermal neutron columns as well as isotopic neutron sources and neutron generators can be used as neutron sources for LSNAA.

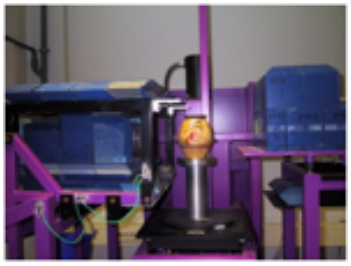


Fig. 3 Measurement of an irradiated vase as a whole

LSNAA has been successfully applied for the analysis of stone samples, entire potsherds of archaeological interest, irregular-shaped bulk historical objects and artefacts as well as in authentication studies of ancient porcelain ware [5].

Concluding remarks

The simplicity of the procedure, the fact that little or no sample consumption occurs during the analytical process and its excellent sensitivity are great advantages that distinguish NAA from other analytical techniques. Therefore, NAA has been established as an important analytical tool and is often considered as the reference technique for comparison of other analytical methods. A great new prospect is the analysis of large volume samples. LSNAA in combination with other techniques, such as neutron imaging and tomography, provides a powerful novel analytical tool with unique new applications in science and technology.

(1) Greenberg R. R.; Bode P.; De Nadai Fernandes E. A., Spectrochimica Acta B, 2011, 66, 193?241
(2) Glascock M. D.; Neff H., Measurement Science and Technology, 2003, 14, 1516?1526
(3) Bode P., J. Radioanal. Nucl. Chem.. 2012, 291, 275?280.
(4) Ellis K.J., Physiological Reviews, 2000, 80, 649-680
(5) Bode P., Activation analysis of large samples, Encyclopedia of Analytical Chemistry, 2008, R.A. Meyers (Ed.), John Wiley Sons Ltd

The New Generation of Neutron Detectors for the European Spallation Source

by Dr. Kalliopi Kanaki †

Introduction

The European Spallation Source ERIC (ESS) is aspiring to become the most powerful neutron source in the world and is currently under construction in Lund, Sweden. It is the first attempt for a European-wide project of this scale in the neutron scattering community. The challenges are plenty, both on the collaborative and the technical front. This contribution is aiming to inform the reader about how the development of a new generation of neutron detectors can tackle the unprecedented requirements the design of ESS neutron instruments imposes. Each instrument class comes with a scientific agenda, whose purpose is to push forward the frontiers of knowledge in a certain field. The high level scientific requirements are broken down to and translated to technical requirements for each of the instrument components, e.g. guides, choppers, sample environment etc. and finally detectors, which register the scattered neutrons from the sample under study. Grouping these requirements, a landscape of clear development directions is forming. A selection of these directions, funded by the BrightnESS - Horizon 2020 grant [1], is briefly presented here: a. high counting rate capability, b. high spatial resolution, c. large coverage area.

The High Counting Rate Frontier

Reflectometry is one of the biggest challenges at ESS. With a power of 5 MW and a proton energy of 2.5 GeV, the incident flux on the sample is anticipated to be of the order of 10^{10} $n/s/cm^2$. With samples reflecting a significant percentage of the incident beam, it becomes apparent that this is the approximate scale of flux that the detector has to withstand. A dedicated detector design, the Multi-Blade, is addressing this challenge. It improves the capabilities of existing reflectometry detectors by a factor 2 in spatial resolution and at least by a factor 10 in terms of rate. It is a ^{10}B-based MultiWire Proportional Counter with several cathode and anode wire planes arranged in the shape of a fan (see Fig. 1). The low incident angle of the neutron on the boron-coated cathodes allows for high detection efficiency and high rate capability, as the incident neutrons are spread over a wide surface area. The position information comes from reading out anode wires and the cathode strips in coincidence. It has been successfully tested at neutron beam lines without saturation up to $1.6kHz/mm^2$ [2] (see Fig. 2).

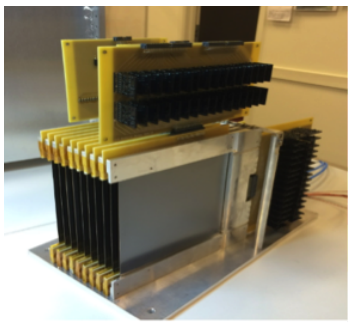


Fig. 1: Side view of the Multi-Blade detector with its front-end electronics. The dark surface is the boron-coated cathode.

†Detector Group, European Spallation Source ERIC

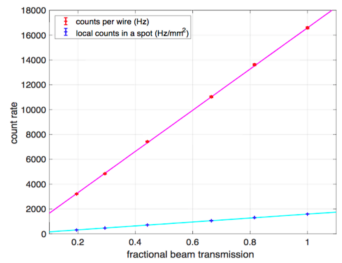


Fig. 2: Count rate vs. beam flux on the Multi-Blade detector.

The High Spatial Resolution Frontier

NMX (Neutron Molecular Crystallography) is the ESS instrument dedicated to protein crystallography with neutrons. It requires a detector spatial resolution down to ca. 200 μm, about 5 times better compared to current detectors with TOF capability. To this end, a Gd-GEM detector with a single converter layer has been developed, which uses the μTPC method to reconstruct the conversion products and reach the required spatial resolution [3] (see Fig. 3).

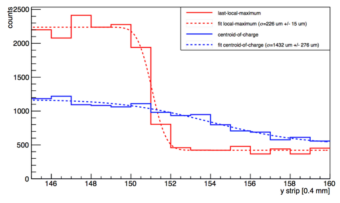


Fig. 3: Distribution of the reconstructed y-coordinate with two different algorithms. In red, the μTPC algorithm results in a spatial resolution of ca. 225μm.

The High Coverage Area Frontier

Chopper spectrometers are traditionally an instrument class that requires some of the largest area coverage (tens of m^2). Given the high price and low availability of ^{3}He gas, the ESS Detector Group and ILL have co-developed the Multi-Grid detector [4], in order to keep the cost under control without compromising the scientific requirements (see Fig. 4).

Fig. 4: A section of the Multi-Grid detector (32 cm x 48 cm). It can be scaled up to the areas required by chopper spectrometry.

The large coverage area is the key characteristic of this ^{10}B detector. This design has been developed over the course of several years, is already scaled up in size with the construction of a 3 m x 0.8 m long unit and has been exhaustively tested at beam lines under realistic neutron beam conditions, both at reactor and spallation sources. Latest tests were performed at the CNCS spectrometer at the SNS spallation source in Oak Ridge, side-by-side with ^{3}He tube detectors (see Fig. 5).

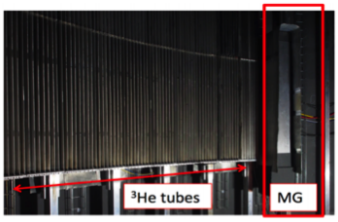


Fig. 5: The Multi-Grid module installed next to the ^{3}He tubes of the CNSC instrument at SNS.

Fig. 6 depicts an equivalent performance between the two detectors [4]. However, the Multi-Grid can provide it only for a fraction of the ^{3}He cost.

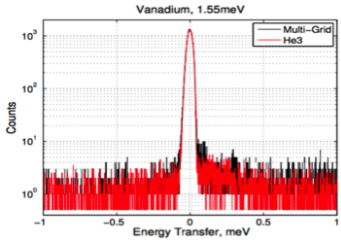


Fig. 6.: Energy transfer spectra derived from ^{3}He tubes and the Multi-Grid detector for a Vanadium sample at 1.55 meV incident neutron energy.

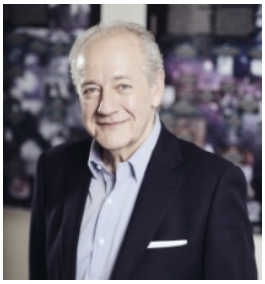
Summary

The detectors of the upcoming ESS instruments are going to look very different compared to other spallation sources. A new era of exciting experimentation is ahead of us. Some of the experimental results of the ESS detector design process were very briefly summarized in this letter. A more thorough presentation can be found in the respective references.

(1) BrightnESS grant: 676548
(2) F. Piscitelli et al., "The Multi-Blade Boron-10-based neutron detector for high intensity neutron reflectometry at ESS", JINST 12, P03013 (2017).
(3) D. Pfeiffer et al., "First Measurements with New High-Resolution Gadolinium-GEM Neutron Detectors", JINST 11, P05011 (2016).
(4) A. Khaplanov et al., "Multi-Grid Detector for Neutron Spectroscopy: Results Obtained on Time-of-Flight Spectrometer CNCS", JINST 12, P04030 (2017).

News

Walter Haelg Prize 2017 awarded to Professor Juan Colmenero



The most outstanding contribution of Juan Colmenero in the field of neutron scattering on complex materials such as polymers or soft matter in general was the creation of a pioneering, unique and robust scientific methodology based on the combination of neutron scattering with different spectroscopic methods and molecular dynamics (MD) simulations. He was one of the first to recognise that in intricate soft matter systems often neutrons alone are not enough to tell us, 'where the atoms are and what they do' and that neutron scattering and MD simulations are natural partners. Taking inspirations from classical polymer, soft matter and condensed matter physics,

he followed an interdisciplinary approach addressing different topics such as the physics of non-crystalline solids, water, polymeric materials, soft matter systems, biopolymers and nano-structured materials. Early on he focused on the different relaxation processes that take place in glass forming polymers. There the special strength of his methodology is evident that is able to cover a huge frequency range and at the same time provides spatial resolution. Major results are: (i) the realization of an analytical relation between the stretching of a relaxation function and the momentum transfer dependence of the corresponding relaxation time for Gaussian relaxation processes; (ii) the observation of quantum rotational tunneling of methyl groups in glassy polymers related to the intrinsic disorder and distribution of local potentials; (iii) the deciphering of the motions in water that cause its dielectric response and their impact on the structural relaxation; (iv) the understanding of dynamic decoupling in thermodynamically miscible but dynamically strongly heterogeneous polymers. During the last years Juan Colmenero focused on the structure and dynamics of more complex soft matter systems such as multicomponent, confined or nanostructured materials. This forced him to extend his strategy to coarse-grained simulations. In this direction, particularly remarkable are the recent and pioneering works dealing with the formation of single-chain functional soft nanoparticles by combining advanced chemistry, coarse-grained simulations and SANS. Juan Colmenero graduated from the University of Navarra, Spain, In 1979 he took a postdoctoral position in the Minsterio de Trabajo, Spain. Thereafter he went to the University of the Basque country in San Sebastian (Spain), where he started as an assistant professor. In 1989 he was appointed full professor at the faculty of chemistry and became the director of the Materials Physics Department. From 1999 to 2013 he was director at the of the ?Donostia International Physics Center? that promotes international collaboration around neutron scattering techniques and provides a meeting point in San Sebastian for scientists from all over the world. From 2001 to 2011 he was the director of the Materials Physics Center (CFM), CSIC-UVP/EHU. Juan Colmenero has significantly contributed to raise the profile of neutron scattering in Spain. He was one of the founders of the 'Sociedad Española de Técnicas Neutrónicas (SETN)' and its first chairman. He is recipient of the 'Xabier María de Munibe' Prize in Science and Technology (1998) of the Basque Parliament, the Euskadi Price (2000) of the Basque government and the gold medal of the Royal Spanish Physical Society (2003).

Neutron Schools

10th ILL School on Neutron Diffraction Data Treatment using the FullProf Suite

We are pleased to announce the 10th ILL Annual School on Neutron Diffraction Data Treatment using the FullProf Suite, to be held in Grenoble at the Institute Laue-Langevin from October 16th to October 20th 2017. Information can be found at the *school's website.*

Scientific scope: FPSchool aims to contribute to the training of scientists in treatment of X-ray and neutron diffraction data. The school is based on intensive hands-on sessions using the computer programs of the FullProf Suite. The lectures and tutorials will provide the essential tools necessary for an efficient use of the FullProf Suite at an intermediate level. In addition to general applications, Rietveld method, multipattern analysis, microstructure determination and refinement, Ionic conduction diffusion paths calculations and magnetic structure determination will be given particular attention.

Lectures: The school lasts 4.5 days: 2 days dedicated to general applications plus 2.5 days dedicated to the analysis of different problems on condensed matter science. The theoretical modules are focalized on different problems that will be complemented with different hands-on sessions for which each participant should bring his/her own laptop with the FullProf Suite already installed.

Participants: The school is intended for PhD students and experienced scientists with a basic knowledge of diffraction techniques and crystallography. As usual, among the participants there will be a mix of different levels of experience in using FullProf. It is recommended, to those who consider themselves as beginners, to read the FullProf manual in advance and even to follow a few of the online tutorials. The maximum number of participants is limited to 30, which ensures the quality of the training provided. The selection of candidates is based on his/her motivation letter and CV (plus a recommendation letter from the thesis supervisor for PhD students). If necessary, balance between laboratories and nationalities will be taken into account. For information, about 1 out of 3 candidates is accepted.

Contact with the editorial board

The provisional editorial board welcomes articles and ideas about the contents of the HENA newsletter from fellow scientists in Greece and abroad. For this purpose please contact:

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