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New Developments of Neutron Activation Analysis Applications

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Abstract

Neutron Activation Analysis is the most sensitive analytical technique used to determine concentration of elements in a sample. Methods of neutron activation analysis have been known for quite a long time, since 1936. However, active development of this method in its technological applications has been started together with recent developments of powerful and safe sources of neutrons, precise and fast gamma detectors, high speed electronics, increased processing ability of the computers and growing demands for industrial processes automation quality control. We discuss as these latest developments influenced performance and utility of different Neutron Activation Analysis techniques and first of all accuracy and stability of analytical results. As a result of the mentioned improvements wide variety of neutron analysis devices has been employed recently for reliable and precise control in cement and coal industry, borehole logging, security monitoring, industrial elemental analysis for production automation and quality control, nondestructive testing. Company RatecLab Ltd develops universal detection system of high resolution gamma spectroscopy for tagged neutron analysis with time-of-light filtering (Skolkovo Grant 2014-2015). Such high resolution detection platform would address the shortcuts and problems associated with conventional detection systems that utilize detectors with relatively low resolution prone to high background problem and slow response time. We perform thorough research of new potential applications of such precise and fast Neutron Activation Analysis system for some unresolved demanding problems of industries such as gas and oil industry. In our opinion there are multiple new possibilities to apply systems with neutron interrogation for industrial production control, nondestructive pipe inspection, neutron radiography etc.

Keywords: Neutron activation analysis; cement production; oil production

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1. Introduction

Neutron activation analysis was developed by G.Hevesy and H. Levi in 1936 following the discovery of neutron by J. Chadwick in 1932 and especially the experiments of F. Joliot and I. Curie in 1934. It has been demonstrated that induced radioactivity can be used for determining the presence of unknown elements in samples. At the same time the first experiments in Neutron Radiography were carried out by H. Kallmann and E. Kuhn.

Thereafter, the development of the method and its applications was defined by progress in its components and instrumentations such as nuclear reactors in the 1940s, scintillation detectors like NaI (Tl) in the 1950s, the semiconductor detectors (Ge, Si, etc.), multichannel analyzer in the 1960s, and computers and relevant software in the 1970s.

The method is based on conversion of stable atomic nuclei into radioactive nuclei by irradiation with neutrons and subsequent detection of the radiation emitted by the radioactive nuclei and its identification. But exactly in the recent years significant and rapid developments in technologies such as the neutron sources and detectors together with still increasing computer power make possible to take the full advantage of this technique and significantly broaden usage of neutron analysis in in different applications.

Even for well-known applications now a lot of different specialized techniques with neutron irradiation have been developed, for instance: neutron powder diffraction (for inorganic functional material), neutron beam diffraction (bio-polymer material), neutron small angle scattering (magnetic/polymer material), neutron radiography (industrial materials, cultural objects, agricultural crops), neutron reflectance measurement (thin-film material), neutron doping (semiconductor), activation analysis (non-destructive inspection).

Classical neutron activation techniques have many properties that make them distinguished for certain types of non-destructive testing like imaging and elemental composition analysis. Neutron as a neutral particle can penetrate deeply even in dense material. Neutrons interact only with nuclei and initiate different nuclear reactions depending from neutron energy. Elastic and inelastic neutron scattering and neutron capture reactions as well as its combination are used for the development of the variety of Neutron Activation Analysis (NAA) techniques such as PGNAA (Prompt Gamma Neutron Activation Analysis), PFNA (Pulsed Fast Neutron Analysis), PFTNA (Pulsed Fast/Thermal Neutron Analysis), API (Associated Particle Imaging).These techniques have big application potential since they could provide data about large number of elements simultaneously and non-destructively together with valuable imaging information.

Last decade of research and development introduced a lot of NAA techniques to the scientific (Parsons et al., 2011), and medical research (Kehayias and Shuang 1993; Maglich &Nalcioglu, 2010), various material analysis applications, ranging from coal (Dep et al., 1998; Sowerby, 2009) and cement analysis (Womble et al., 2005), borehole logging (Nikitin & Bliven, 2010) to various explosive detection schemes (Vourvopoulos & Womble, 2001; Aleksandrov et al., 2005; Lanza, 2006), such as detection of threats in cargo containers and vehicles (Barzilov & Womble, 2003, Reber etal., 2005; Koltick et al., 2007) and land mine detection (Womble et al., 2002; Holslin et al., 2006).

In the present work we observe some of the latest developments in the basic components of neutron interrogation methods and discuss how these developments influence that technique potential applications.

2. Neutron sources development

Some radioisotopes, nuclear reactor, or accelerator-based devices can be used as neutron sources. Radioisotopes used as the neutron sources include ^{252}Cf, ^{239}Pu, ^{241}Am, and others. Radioisotope sources of neutrons are cheaper and smaller then neutron generators but neutron yield of these sources is decaying with time and typically limited to a total intensity of 10^7 n/s. Also radioactive sources capable of producing high neutron flux contain hazardous quantities of radiation requiring many safety considerations.

Neutrons can be produced with neutron generators (NG) by the deuterium–deuterium (DD) (with energy 2.5 MeV) or deuterium–tritium (DT) (with energy 14 MeV) reactions. Unlike radioactive sources, NGs contain no radioactivity (except those based on the DT reaction) what makes them inherently safe when it turned off. A limiting factor of these generators consists in the erosion of the neutron producing target which has their typical lifetime of the order of few thousand hours. Sealed tube neutron generators are produced by Thermo Fisher Scientific, Schlumberger, Baker Hughes, EADS SODERN in France, and VNIIA in Russia.

Neutron yield, neutron producing target lifetime and portability of NGs have been significantly improved within the last decade. Nowadays neutron yield of NGs produced by VNIIA raised up to 10^{11} neutron/s (VNIIA, http://www.vniia.ru/eng/ng/index.html). Thus portable NGs have found many applications in industrial process analysis and control.

The associated particle imaging (API) - one of the very promising and useful for many applications approach has been recently brought out of the laboratory with the development of transportable associated-particle sealed-tube neutron generator (APSTNG). In these sealed tubes each neutron is emitted simultaneously and in opposite direction with the alpha particle, which detection by a position sensitive detector determines the direction of the alpha particle and thus the direction of the outgoing neutron. Gamma rays are produced when this “tagged” neutron interacts with materials of interest. Produced characteristic gamma rays are detected in coincidence with the alpha particle, generating of a neutron time-of-flight (TOF) spectrum. The TOF allows to determine the position of the neutron interaction. Thus not only elemental composition analysis but also 3D imaging of the elements position inside the object is possible (Koltick et al., 2009). Such API based systems can be applied and as a scanning tool to detect explosives, drugs or nuclear-based contraband in cargo or vehicle, for different purposes in biomedical research or for malignant tumors in the human body diagnostics. Development of the APSTNG makes possible to use such precise and elaborate method for variety of problems with still big potential to be realized.

3. Gamma ray detectors development

For implementation of the precise and reliable elemental analysis high resolution gamma spectroscopy is required. Gamma spectrometers gives information both on energy and intensity of the radiation emitted from the source. The main component of gamma spectroscopy system is gamma detector which physical characteristics and limitations influence parameters of the entire system.

There are a lots of requirements for gamma ray detectors suitable for NAA based system:

- It must operate in environment where neutrons and gamma rays are present,
- The detector material must have a high atomic number,
- The detector must provide the energy resolution that allows resolving peaks of interest,
- Neutron induced gamma ray response from the sample shouldn’t interfere with the response from detectors material.

Selection of the suitable detectors with needed characteristics considering the costs is a complicate task for the development of any particular NAA system based on their required properties and application field.

Until recently sodium iodide (NaI(Tl)) was mainly the most popular scintillator for such NAA detectors system. However, it is lacking in some desirable properties like the speed of signal, energy resolution and effective atomic number. As a result, a number of new scintillators are emerging that improve in one or more of these categories.

The new developments in inorganic scintillators for gamma ray detection have been made during last decade. For example lutetium orthosilicate or LSO (Lu_2SiO_5:Ce) with unique combination of high luminous efficiency, high density, and reasonably short decay time. Several recently discovered materials (such as $LaBr_3$:Ce) possess energy resolution that approaches level of direct solid state detectors.

Another solution for high resolution spectroscopy is a high purity germanium detectors (HPGe) which represent a solid-state semiconductor detector that require liquid nitrogen or mechanical cooling subsystems. It has superior energy resolution comparing to scintillation detectors (see Fig.1). The speed of the HPGe signal collection is another superior parameter for high count rate conditions and applications that require good timing resolution (Cooper & Koltick, 2001).

In conclusion it should be noted that NAA systems equipped with high resolution detectors require fast data acquisition electronics for the appropriate detectors signal processing.

One of such high-resolution detection system for gamma radiation spectroscopy with neutron time-of-flight filtering with HPGe detectors been developed by Dioszegi et al (2014). Such system can be used for mentioned above API - Associated Particle Imaging technique. API method as a superior tool for accurate gamma signal measurement and imaging requires high-resolution gamma radiation spectroscopy only possible with new advances in gamma ray detectors and acquisition electronics.

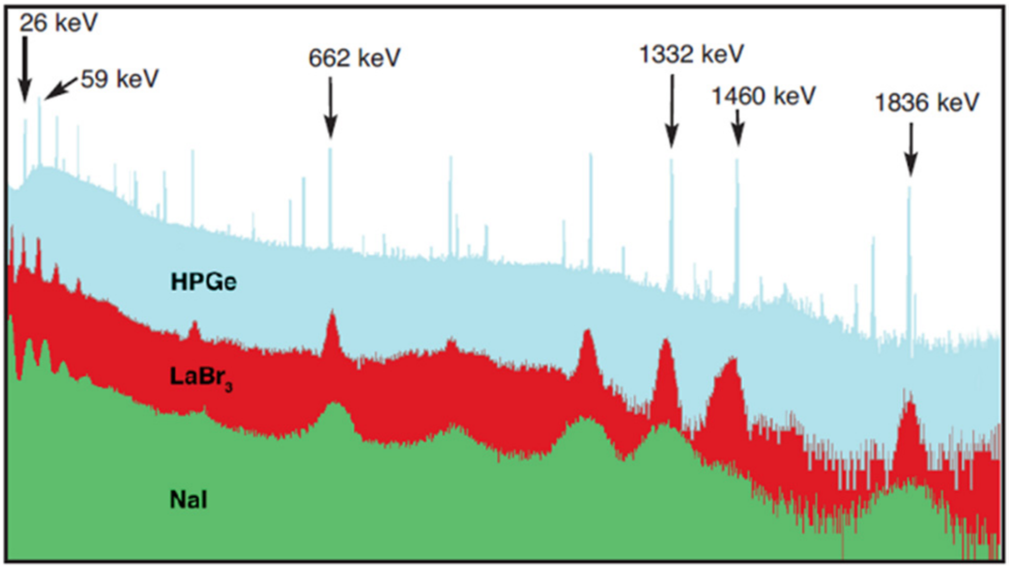


Fig. 1. Comparison for $LaBr_3$(Ce), NaI(Tl), and HPGe spectra (ORTHEC)

4. Neutron interrogation for medicine

Neutron based technologies can be very effectively applied for the cancer diagnostics. While mammography (x-ray imaging) which is presently mainly used for breast cancer diagnostic could not determine whether the tumor is benign or malicious. For the confirmation of the diagnosis the invasive biopsy is needed. The other disadvantage is, that the radiologists working on the interpretation of the mammography images need a long and expensive training before they are able to set a good diagnosis.

In recent years a growing body of evidence has indicated that many trace elements play an essential role in a number of biological processes. Statistically significant differences from the normal distribution of potassium, rubidium, magnesium, calcium, copper, zinc, iron, selenium, manganese, and other essential elements have been reported to occur in patients with various forms of cancer (Danielson and Steinnes 1970, Mulay et al. 1971, Schwartz and Fink 1974, Valcovic et al. 1980). Changes of trace element concentrations in human tissue may be a precursor to malignancy in several other organs like brain or prostate several cancers including breast, lung and colon.

Duke University researchers are developing Neutron Stimulated Emission Computed Tomography (NSECT) that will be able non-invasively mapping the concentration of any isotope in any selected 3-D volume of the body (Kapadia et al., 2008). Thus breast cancer could be diagnose earlier by imaging the relative concentrations of trace elements in breast tissue and detecting the subtle differences in chemical composition between benign and malignant tissue. The practical application of such technology could extend from very early breast cancer screening or liver involving iron overload such as hemochromatosis, as well as unusual levels of copper such as Wilson's disease to large variety of biological, medical and pharmaceuticals fundamental research.

Another approach is based on oxygen concentration measurement developed by California Science & Engineering Corporation (CALSEC). The differential femto oximetry (DFO) method is utilized the fact that cancerous tumors have different oxygen content than healthy tissue. Therefore, by measurement the oxygen concentration difference between a tumor and the adjacent healthy tissue malicious tumor can be indicated. A compact probe Oncosencor designed by CALSEC emits 2 or more pencil beams of femto-neutrons (fast neutrons of femto-meter wave-length) aimed to provide "needleless biopsy" for breast cancer diagnostics (Maglich & Nalcioglu, 2010).

It can be noted that neutron based techniques for cancer diagnosis are at an advanced stage of development now and have a lot of potential be practically implemented in practice.

5. Associated Particle Technique – from cargo scanning for dangerous and illicit objects to the medical and biological studies

Some extensive studies have been done in a field of application API technique for the explosives and illicit objects detection (Bystritsky et al., 2003; Perot et al., 2007; Bystritsky et al., 2008). Research shown that API technique as compared with other detection techniques (other methods of activation analysis, X-rays, nuclear quadruple resonance etc.) demonstrates a lot of advantages such as:

- 3D location and imaging of the object;
- Elemental composition analysis of the matter;
- High penetration of fast neutrons;
- Higher sensitivity (effect/background ratio is 200 times better than without "tagging").

API system with APSTING as a neutron source can be used alone or as supplement for second inspection after the x-ray scanning for the suspicious object verification (see Fig.2).

API system of elemental analysis could be offered for animal physiologist as a tool to monitor the growth of an animal in response to new genetic, nutritional and pharmacologic methods for livestock improvement. Possibility to apply APSTING for analysis of total body carbon (TBC), total body nitrogen (TBN) and total body oxygen (TBO) for protein, fat and water in vivo determination has been demonstrated by Mitra et al. (1995, 2013). The radiation dose equivalent delivered due to neutrons was calculated to be approximately 0.03 mSv.

It is clear that search for the potential API technique application is not complete yet and has great future in medicine, biological studies, security and other non-resolved yet industry problems.

Fig. 2 Cargo inspection portal based on API technology (Perot et al., 2007)

6. Neutron interrogation for radiography and elemental analysis

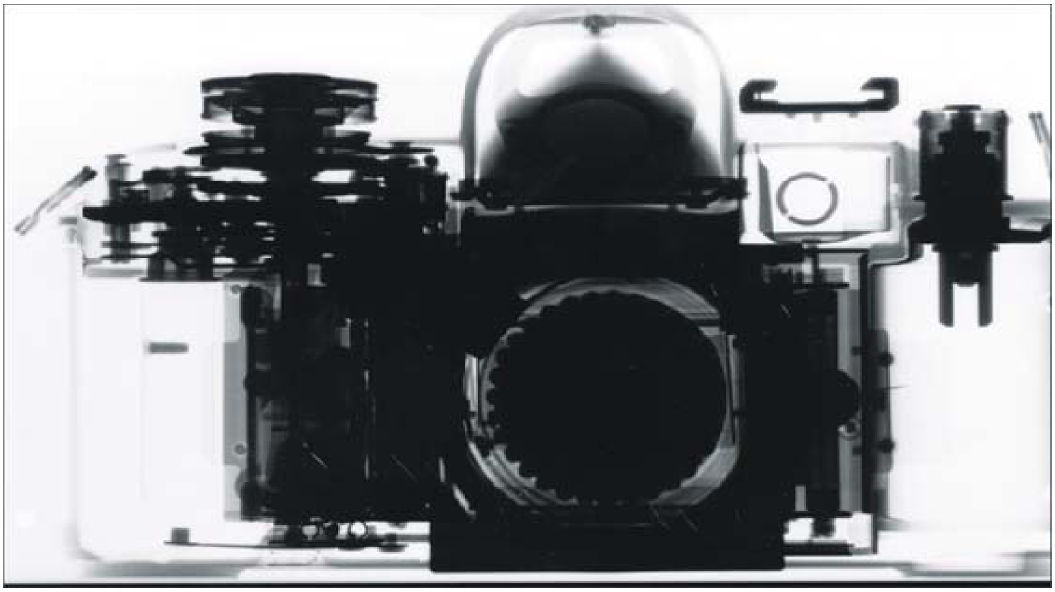
Neutrons unique characterizes makes them suitable for a number of tasks impossible for any other methods of non-destructive analysis like x-ray. Neutron easy penetrate heavy elements such as lead, titanium etc. At the same time they are sensitive to light element such as hydrogen. It makes neutron scanning very promising technique for solving some industrial problem. On of such complicate problem is detection of hydrocarbon deposit and corrosion in industrial pipes.

Hydrocarbon deposits can cause large economic losses for different industries, such as refineries or petrochemical plants. Wax blockage of pipeline could lead to great economical losses therefore pipes need regular inspection. Although detection of such hydrocarbon deposits represents a big problem. Usual inspection techniques such as gamma or x-ray radiography could not help since density and atomic number of pipe walls are much higher than that of deposit. Attenuation of gamma rays mostly in iron walls makes contrast of radiography very poor. Ultrasound methods require preparation of the pipe surface.

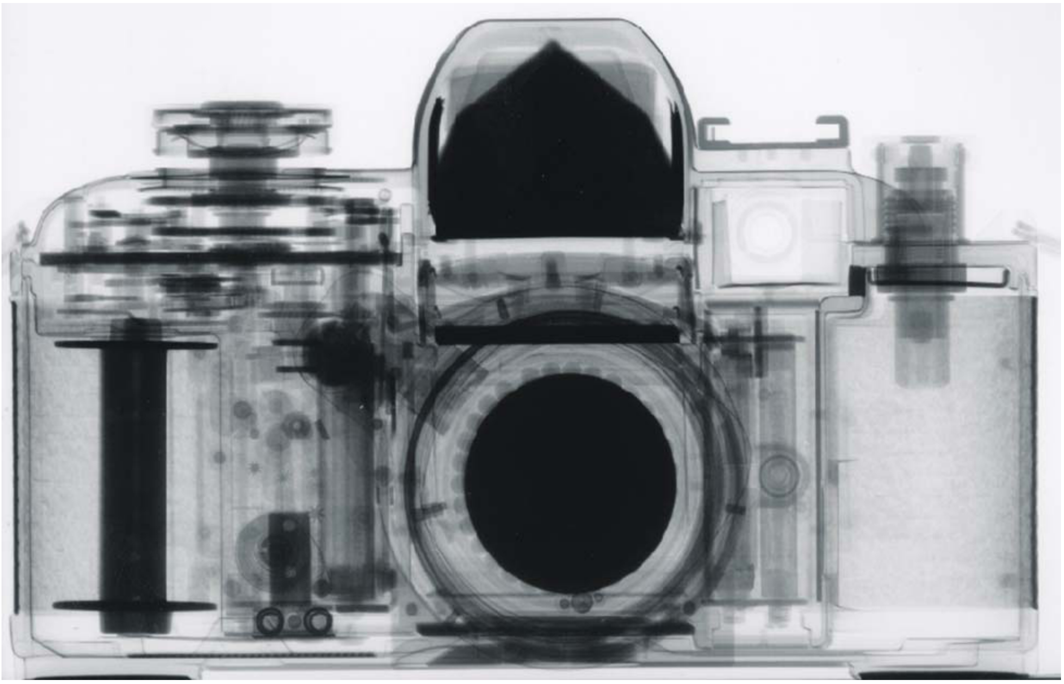
Application of the neutron back diffusion and neutron capture gamma rays analysis for measuring of wax deposition and corrosion in pipelines has been studied by Abdul-Majid (2013). Neutron capture gamma ray measurement was successful for determination iron pipe wall thickness and organic scale accumulation inside the pipes.

Another approach for on-line inspection of scale and corrosion was studied by Fujine et al (1983), who used the video image processing system for the real-time neutron radiography. Neutron radiography is very powerful technique with unique properties due to it sensitivity to the light elements which are impossible to be revealed by gamma or x-ray scanning (see Fig.3).

In conclusion it should be noted that any of such neutron interrogation system can be made portable, could work on hot, cold or insulated pipes. They can be utilized as for deposit composition and pipe wall imaging and characterization, as for visualization of multiphase flows in tubes or detecting certain elements in the flow.



a) X-Ray Radiograph



b) Neutron Radiograph

Fig.3. Difference between X-ray and Neutron radiographs of a 35 mm film SLR camera. Dark elements in the x-ray radiographs are metal components they are almost transparent for neutrons. While in the neutron radiograph dark components are plastic components which are almost transparent to x-rays (McClellan Nuclear Research Center, Neutron Radiography).

7. RatecLab: Development of Universal Spectroscopic Platform

STC RATEC since its creation in 1991 has been developed different system based on neutron activation analysis for detection of explosives, radioactive threats and Special Nuclear Material with Thermal Neutron Analysis for passenger luggage inspection. STC RATEC has created a daughter company in the framework of Skolkovo Foundation- RatecLab. Company RatecLab develops universal detection system of high resolution gamma spectroscopy for tagged neutron analysis with time-of-light filtering (Skolkovo Grant for project NEWTRAN). Such high resolution detection platform will address the shortcuts and problems associated with conventional detection systems equipped with detectors with relatively low resolution prone to high background problem and slow response time. As a result the average time of inspection of large cargo container with the neutron interrogation system based on such detection platform expected to be only 8 min and imaging space resolution about 5 mm.

The neutron interrogation system for detection of the explosives and other illicit objects based on such high resolution detection platform will address the main expectations of the market:

- Universality, i.e. the ability to detect all four major types of dangerous material – weapons (both firearms and cold weapons), explosive materials, radiological materials (isotopes), and Special Nuclear Materials for Mass Destruction Weapons (weapons-grade uranium, plutonium)
- Efficiency, i.e. correspondence with the international standards on the reliability of the inspection results and grade of false alarms
- High operation speed (e.g. less than 10 min per standard 40ft sea container)
- Adjustability, i.e. the possibility of being installed in various environments, particularly where there is limited space.

Furthermore we are planning to test the developed detection platform for non-destructive industrial analysis for cement production control and for multiphase flow metering in oil exploration.

8. Conclusion

General advances in science and technology of neutron generation and radiation detection and measurement makes possible to transfer new ideas and approaches of this powerful technique from the laboratory into the everyday life.

Pushing by new technological challenges the scope of applications of neutron interrogation methods is continue to grow. Neutron imaging technique can be a good illustration for that: it is becoming nowadays indispensable analytical tool for Hydrogen Economy and have high potential to be a basis for one of the most advanced Spectroscopic Medical Imaging technique or Neutron Stimulated Emission Computed Tomography.

It can be said that at the present time neutron methods experience the time of renaissance and we can expect a lot of new discoveries and applications in that field.

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