

THESIS FOR THE DEGREE OF LICENTIATE OF ENGINEERING

# **GAMMA SPECTROMETRIC MEASUREMENTS OF NUCLEAR MATERIAL FOR NUCLEAR FORENSIC PURPOSES**

**Study of signatures in  $^{241}\text{Am}$  sources and evaluation of uranium categorization  
with low-resolution instruments**

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CHALMERS UNIVERSITY OF TECHNOLOGY

Gothenburg, Sweden, 2016

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## Abstract

Nuclear forensics is a scientific discipline that aims to aid in criminal investigations concerning illicit use of nuclear and other radioactive material. The discipline aims to find signatures within the nuclear material that can help to find the attribution, or the origin and intended use of the seized material. Examples of signatures are isotopic composition, age and trace element content.

The work presented in this thesis sheds some light upon limitations and possibilities in measurements of radioactive material using hand-held gamma spectrometric instruments commonly used by first-responders or in early stages of a nuclear forensic investigation, i.e. in the detection and identification of the material at e.g. the finding place. One part of the thesis evaluates the capability of categorizing uranium using low-resolution gamma spectrometry and gives a plausible explanation to why the categorization may become erroneous. The second part investigates what kind of information that can be extracted from a simple high-resolution gamma spectrometric measurement of a strong  $^{241}\text{Am}$  source.

The results show that categorizing uranium using low-resolution gamma spectrometry is a challenging task, due to the characteristics of these instruments. To make trustworthy categorizations of uranium based on low-resolution measurements, the properties of the sample as well as possible shielding needs to be known. The results of the investigation of  $^{241}\text{Am}$  sources show that there are a number of promising signatures, such as radioactive and stable impurities and age, which may be used to identify a source when other information about the source is unavailable.

Keywords: uranium, gamma spectrometry, americium, signatures, nuclear forensics, abundance

# LIST OF PUBLICATIONS

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This thesis is based on the work contained in the following papers:

## Paper I

Vesterlund, A., Ulvsand, T., Lidström, K., Skarnemark, G., Ekberg, C., Ramebäck, H., On the categorization of uranium materials using low resolution gamma ray spectrometry, *Appl. Radiat. Isot.*, 72 (2013) 54–57.

## Paper II

Vesterlund, A., Chernikova, D., Cartemo, P., Axell, K., Nordlund, A., Skarnemark, G., Ekberg, C., Ramebäck, H., Characterization of strong  $^{241}\text{Am}$  sources, *Appl. Radiat. Isot.*, 99 (2015) 162–167.

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# 1 ABBREVIATIONS

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ANSI	American National Standards Institute
DU	Depleted Uranium
GC-MS	Gas Chromatography-Mass Spectrometry
HEU	Highly-enriched Uranium
HPGe	High Purity Germanium
ICP-MS	Inductively Coupled Plasma-Mass Spectrometry
ITWG	Nuclear Forensics International Technical Working Group
LEU	Low-enriched Uranium
MCNPX	Monte Carlo N-Particle eXtended
NFKMS	Nuclear Forensics Knowledge Management System
NNFL	National Nuclear Forensics Library
NPT	Non-Proliferation Treaty
NU	Natural Uranium
ROI	Region of Interest
SEM/EDS	Scanning Electron Microscope with Energy Dispersive X-ray Spectroscopy
SIMS	Secondary Ion Mass Spectrometry
TEM	Transmission Electron Microscopy
TIMS	Thermal Ionization Mass Spectrometry
VGSL	Virtual Gamma Spectrometry Laboratory
XRD	X-ray Diffraction
XRF	X-ray Fluorescence





## 2 INTRODUCTION

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Nuclear material has worldwide been under strong control for almost 60 years. The safeguards organization under IAEA has the main responsibility to ensure that states maintain accountability of nuclear material. Even so, there have been incidents where nuclear material has been found out of regulatory control [Wallenius et al., 2006; Wallenius et al. 2007]. The main part has been smuggling incidents but there have also been incidents where theft of nuclear material has been revealed. Whereas the safeguards organization is meant to prevent incidents involving nuclear material, there is also a need for imposing sentences upon individuals who have handled nuclear material illegally [UNSCR 1540, 2004]. The purpose of legal proceedings may be twofold; deterrence and retribution. The deterrence is directed against both individuals as well as against states. The complex production of nuclear material makes state involvement and a state's knowledge of the presence of nuclear material production inevitable. A confiscation of nuclear material found out of regulatory control would imply that a state has failed to follow international law concerning nuclear material. This fact would reinforce a state's will to control nuclear material and thereby deter from both negligence and proliferation. To help in these legal proceedings there has been a need for forensics that focuses on the information that can be provided from the nuclear material, to complement traditional forensic evidence.

Nuclear forensic science is a scientific discipline that has been developing for the last 20 years or so. The aim of nuclear forensics is to aid criminal investigations of nuclear material or other radioactive material to find the attribution, or the origin and intended use of the seized material. Commonly, the investigated material is nuclear material such as uranium and plutonium, but also radioactive sources may be subject of investigation. Furthermore, there are other radionuclides, such as  $^{241}\text{Am}$ , that are fissionable and hence sometimes referred to as alternative nuclear material [IAEA, 2002]. Many of the methods used in nuclear forensics are based on methods used in other disciplines such as age dating and lanthanide analysis which are methods commonly employed in geology [Cheong et al., 2015; Lobato et al., 2015]. One difference in nuclear forensics compared to geology is the need for a different time scale for age dating in nuclear forensics. Whereas the measured ages in geochronology may be millions or billions of years, the age of nuclear material is less than 80 years. This time scale requires other chronometers to provide low measurement uncertainties.

The objective of the work presented in this thesis has been to shed some light upon limitations and possibilities in measurements of radioactive material using hand-held gamma spectrometric instruments commonly used by first-responders or in early stages of a nuclear forensic investigation, i.e. in the detection and identification of the material at e.g. the finding place. The first paper shows some of the difficulties in categorizing uranium using low-resolution gamma spectrometry and the possibility of making erroneous decisions based on the measurements. The second paper shows what kind of information that can be extracted from a simple high-resolution gamma spectrometric measurement of a strong radioactive source. This information could be included in a National Nuclear Forensics Library to help identify a radioactive source found out of regulatory control.



## 3 BACKGROUND

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### 3.1 Political history of nuclear development

In 1938 Otto Hahn and Fritz Strassmann irradiated uranium with neutrons and discovered that one resulting entity from the irradiations was barium, but they could not explain how this was possible [Hahn and Strassmann, 1939]. Shortly after, Lise Meitner and her nephew Otto Frisch devised the theoretical explanation that the neutron irradiation split the uranium atom into two nuclei of roughly equal size and that energy is released in the process [Meitner and Frisch, 1939]. Fission was discovered and shortly after this it was found that the neutrons produced in the fission could cause a chain reaction [Zinn and Szilard, 1939]. In 1940 Rudolf Peierls and Otto Frisch wrote an article on the possibility to build “super-bombs” using uranium enriched in  $^{235}\text{U}$  [Arnold, 2003], and July 16 1945 the first nuclear test, the “Trinity” test, was conducted. The test was followed by the atomic bombs over Hiroshima and Nagasaki shortly after. Soon, technical details on the construction of the atomic bomb leaked to the Soviet Union, and when the Soviet Union had implemented the nuclear weapons technology in 1949, when the first Soviet test was conducted, the nuclear arms race became a part of the Cold War.

The IAEA was established in 1957 after the “Atoms for Peace” initiative by the US president Dwight D. Eisenhower to promote peaceful use of nuclear energy and to prevent nuclear material to be used for military purposes [Fischer, 1997]. The idea was that the IAEA would serve as a bank for nuclear material. A safeguards organization was created to ensure that nuclear material was not used for military purposes. However, the Cold War prevented the implementation of the IAEA as a nuclear material protector. Instead, while the nuclear technology intended for peaceful purposes spread across the world, so did the nuclear weapons technology. For example, the plutonium used for the Indian nuclear weapons programme was produced in a research reactor supplied by Canada [Fischer, 1997]. By 1968, five countries around the world had nuclear weapons technology and, furthermore, conducted nuclear weapons tests. It became clear that the spread of this knowledge and technology should be stopped. In 1970 the Nuclear Non-Proliferation Treaty (NPT) entered into force.

The NPT can be described as having three pillars; non-proliferation, disarmament, and peaceful use of nuclear technology. The first part, non-proliferation, obligates the nuclear weapons states not to share nuclear weapons or technology related to nuclear weapons to states that do not have nuclear weapons. On the other hand, non-nuclear weapons states are obligated not to receive or develop nuclear weapons technology. Also, non-nuclear states are obligated to accept IAEA safeguards to verify that the nuclear technology within the state is used for peaceful purposes. The safeguards organization maintains credible assurance that nuclear material under safeguards is used for peaceful aims and is not converted into nuclear weapons [NPT/CONF.2015/13, 2015]. The second pillar, disarmament, implies that the nuclear weapons states are obligated to pursue the complete disarmament of nuclear weapons, and the third pillar gives the states the right to develop and use nuclear technology for peaceful purposes, e.g. nuclear energy [Reed and Stillman, 2009]. After the implementation of the NPT, still a few states have developed and tested nuclear weapons. After the disassembly of the Soviet Union there were suddenly three new nuclear weapons states. However, these three states, Belarus, Kazakhstan and Ukraine, soon transferred their stock of nuclear weapons to the Russian Federation [Reed and Stillman, 2009]. In the aftermath of the breakup of the Soviet Union, seizures of illicit radiological and nuclear material at borders started to increase rapidly. IAEA recorded more than 2150 incidents of illicit trafficking of radioactive material between 1993 and 2011. Of these, more than 400 cases concerned depleted uranium (DU), natural uranium (NU) or low-enriched uranium (LEU) and about 16 concerned highly-enriched uranium (HEU) and plutonium [Hutcheon et al., 2013].

To handle events with nuclear material and other radioactive material out of regulatory control, the IAEA has established a nuclear security programme. This programme focuses on prevention and detection of and response to criminal or other unauthorized acts involving nuclear or other radioactive material [IAEA, 2011]. The IAEA recommends each state to implement a nuclear security infrastructure to protect nuclear and radioactive material within its borders. The state should also have the ability to “prevent, detect and respond to nuclear security events” [IAEA, 2015]. The preventive measures involve deterrence, information security, and trustworthiness of personnel by the implementation of a “nuclear security culture”. The detection measures should involve detection by instruments as well as information alerts. The response measures involve the actions that follow a detected nuclear security event and includes notification and activation of all relevant authorities including the initiation of investigations concerning the event. For the purpose of both deterrence and response, nuclear forensics plays an important part.

### 3.2 Nuclear forensics

Nuclear forensics is a relatively new scientific discipline. The aim of nuclear forensics is to find the attribution of unknown nuclear and other radioactive material.<sup>1</sup> The attribution assessment is used to determine the origin of the material, the intended use and the responsible individuals connected to the material [Hutcheon et al., 2013]. Methodology in nuclear forensics includes measurements of radioactive nuclides as well as stable elements. Some of the methods, such as age dating and lanthanide patterns, are also used in geology and archaeology. However, a major difference between nuclear forensics and, for instance, geology is the need for quality assurance to meet the high legal and scientific scrutiny [Leggitt et al., 2009]. Therefore a considerable amount of the conducted research focuses on improving the measurement precision and obtaining smaller uncertainties [Williams et al., 2014]. Nuclear forensics often combines laboratory methods used for ordinary analysis of nuclear and other radioactive material and interpretation of the analyses to provide technical conclusions about, for example, seized illicit nuclear or radioactive material [Kristo and Tumey, 2013].

A nuclear forensic investigation can be divided into three parts [IAEA, 2006]:

#### - Categorization

The IAEA report “Nuclear Forensics Support” (2006) states that “*categorization is used to address the threat posed by a specific incident*”. The aim of the categorization is to identify the safety risk to first responders and to the public, as well as to evaluate whether the incident is part of criminal activity and/or a threat to national security. One example of categorization may be the interception of uranium at a border control. The categorization done at this stage could include gamma spectrometric measurements of the found material. Calculating the enrichment of the uranium would provide information to how to proceed with confiscation or even if a crime has been committed.

#### - Characterization

Characterization on the other hand handles the determination of specific characteristics of the material. Tab. 1 lists some useful techniques and methods and gives a recommended sequencing of analysis to give the most valuable information early in an investigation [IAEA, 2015]. The table is a

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<sup>1</sup> The IAEA states that: “*Nuclear forensics is the examination of nuclear or other radioactive material, or of evidence that is contaminated with radionuclides, in the context of legal proceedings under international or national law related to nuclear security. The analysis of nuclear or other radioactive material seeks to identify what the materials are, how, when and where the materials were made, and what their intended uses were*” [IAEA, 2015].

recommendation from the Nuclear Forensics International Technical Working Group (ITWG), which is an association of nuclear forensics practitioners. Important nuclear forensic characteristics include isotopic and elemental composition and physical characteristics. The isotopic composition analysis can be performed using gamma or alpha spectrometry or any of the mass spectrometric techniques depending on sample size [Ramebäck et al., 2012]. The isotopic composition provides information about the intended use of the nuclear material but can also reveal if the material is reprocessed [Zsigrai et al., 2015]. Elemental composition, or impurity measurements, is the measurement of remaining metal impurities and can be used to explain the production process of the nuclear material or the geographical origin of the source material [Healy and Button, 2013; Varga et al., 2010]. Physical characterization is used to tell the grain size or the chemical phase of a material and is mainly performed using surface characterization techniques such as scanning electron microscopy (SEM) and x-ray diffraction (XRD) but also other, more basic techniques, such as weighing and density measurements [Sweet et al., 2013]. Another useful characteristic is the age of a material, i.e. the time that has passed since the last separation. The idea of age dating is to investigate the relation between a mother nuclide and the ingrowth of a daughter nuclide [Eppich et al., 2013; Gehrke and East, 2000; Nygren et al., 2007; Ramebäck et al., 2008; Varga et al., 2011]. The principles of age dating is explained in Chapter 4.

Table 1 Suggested priority for common characterization methods [IAEA, 2006].

Techniques/Methods	24 h	One week	Two months
Radiological	Estimated total activity Dose rate ( $\alpha$ , $\beta$ , $\gamma$ , n) Surface contamination		
Physical	Visual inspection Radiography Photography Weight Dimensions Optical microscopy Density	SEM/EDS XRD	TEM (EDX)
Traditional forensics	Fingerprints, fibres		
Isotope analysis	Gamma spectrometry Alpha spectrometry	Mass spectrometry (SIMS, TIMS, ICP-MS)	Radiochemical separation
Elemental/chemical analysis		ICP-MS XRF Assay (Titration, IDMS)	GC-MS

#### - Nuclear Forensic Interpretation

Besides the analysis of nuclear and other radioactive material, another important part of nuclear forensics is the interpretation of the analytical results. Nuclear forensic analysis can be divided into two groups; comparative and predictive analysis [Hutcheon et al., 2013]. A measurement technique or a measurand can be used both for comparative and predictive analysis, depending on the question. The comparative analyses can be used to answer the questions “Do these materials have the same origin?” or “Does this material correspond to any material in the database or any material we have knowledge about?” The predictive analyses can, on the other hand, be used to explain the origin of the material and the intended use. So, for example, the isotopic composition can be used to compare

different samples to see whether it is likely that they are coming from the same batch, but the composition can also be used to explain the intended use of the material and possibly the production process. Thus, the nuclear forensic interpretation is used both for linking materials and events to each other, and for the determination of the intent of the nuclear security event.

### 3.2.1 National Nuclear Forensics Libraries

The IAEA has recommended that each state should establish a national register of nuclear material and radioactive sources to ensure that nuclear material and radioactive sources are identifiable and traceable, or when this is not practicable, ensure that there are alternative processes for identifying and tracing sources [IAEA, 2004; IAEA, 2015]. Nuclear Forensics Knowledge Management Systems (NFKMS) is a concept that handles these issues, ranging from subject matter experts to databases containing information regarding domestic nuclear and other radioactive material. These are sometimes called National Nuclear Forensics Libraries (NNFL). The purpose of the library is to provide information to determine the origin and history of radioactive or nuclear material which has fallen out of regulatory control, to support nuclear security and non-proliferation. A library can be used to determine the likelihood that, e.g. an orphan source or seized nuclear material is originating from the state where it is found or whether it belongs to another state [IAEA, 2015]. The library may contain analysis information that can be used for comparison with data from analyses in nuclear forensic investigations, as well as information from manufacturers. It is also possible to include archive samples of different sources in the library to facilitate ad hoc comparisons of characteristics of an investigated material and samples in the archive [Wacker and Curry, 2011].

## 3.3 Methodology

### 3.3.1 Gamma spectrometry

Gamma spectrometry is a non-destructive measurement method for gamma emitting radionuclides. The instruments used for gamma spectrometry can be divided into low- and high-resolution instruments. Low-resolution instruments such as NaI(Tl) scintillation detectors [Knoll, 2000] are commonly used as first-responder or customs instruments, but the ability of these instruments to identify radionuclides has been shown to be unsatisfactory [Blackadar et al. 2003; Nelson et al., 2011; Pibida et al., 2004]. This is mainly due to the low-resolution capability. The instrument is not able to separate peaks which are close in energy, which makes the instrument a blunt tool for identification and requires highly qualified users. However, this is not always enough since the resolution often prohibits even manual identification. High-resolution instruments, high purity germanium detectors (HPGe) [Knoll, 2000], do not have this problem. A number of publications have put forward different identification algorithms but the problem seems to remain [Estep et al., 1998; Hofstetter et al., 2008; Sprinkle Jr et al., 1997]. An advantage of low-resolution instruments is that they operate at room temperature as opposed to HPGe detectors which require cooling. The need for cooling in HPGe detectors somewhat limits its flexibility. Another advantage of the low-resolution detectors is that the acquisition is fast due to the high measurement efficiency since e.g. NaI(Tl) detectors can be produced with very high volume compared to HPGe detectors.

#### 3.3.1.1 *Automatic evaluation algorithms*

Many hand-held gamma spectrometers provide the possibility of automatic identification of the measured radionuclides. Furthermore, some instruments provide the possibility of automatic categorization of nuclear material. There are different approaches to automatic identification algorithms. One common approach is template matching where a measured spectrum is compared to a library of spectra stored in the instrument [Burr and Hamada, 2009]. Another approach is region of

interest (ROI) monitoring, where regions of interest are monitored for elevated count rates. [Blackadar, 2005] One problem with template matching that may arise is that the sample characteristics such as density and sample composition may not correspond to the characteristics of the template spectra. For example, the measurement efficiency vs. energy for a dense sample is substantially different from a water sample so the peak proportions may be different which could lead to misidentifications. Another problem is the libraries stored in the instrument. The size of the library is a trade-off. If the library is too small there is a possibility that the instrument is asked to identify a radionuclide that is not present in the library. On the other hand, if the library contains too much information there is a possibility that there are several feasible options which leads to misidentification [Blackadar, 2005].

### 3.3.1.2 Simulations

It is possible to simulate the behaviour of a gamma detector or a measurement of a specific radionuclide using a Monte Carlo technique. In **paper I** a simulation software, Virtual Gamma Spectrometry Laboratory (VGSL) was used to construct detector response functions depending on shielding and sample material [Plenteda, 2002]. VGSL uses a modified version of MCNPX as transport simulation engine [Waters, 2002].

### 3.3.2 Impurities

Whenever a radioactive source or nuclear material is produced, elements different from the wanted radionuclide are present [Peńkin et al., 2015; Gehrke, 1998]. Some of the elements originate from the raw material [Švedkauskaitė-LeGore et al., 2007], some are introduced as contaminants during the manufacturing process [Healy and Button, 2013] and some elements are added as additives to enhance the performance of the material [Cudrnak and Necas, 2011] or added simply as support material. The impurities can be both radioactive and stable. These impurities can be used for characterization of a material, both for comparative analyses and predictive analyses. In **paper II**, the gamma signature that originates from the neutron activation of most probably support material in  $^{241}\text{Am}$ -sources are investigated.

Neutron sources are constructed in a way that the alpha particle from an alpha-emitting radionuclide such as  $^{241}\text{Am}$  or  $^{210}\text{Po}$  reacts with a low-Z element, commonly beryllium, and a neutron is emitted from the low-Z element nucleus. For low-Z elements the Coulomb barrier is sufficiently small to make a nuclear reaction possible [Choppin et al, 2013]. When the formed atom deexcites or decays, gamma radiation is emitted. There have been a number of studies on alpha particle induced gamma rays from nuclear reactions in light elements [Lappalainen et al., 1983; Sastri and Schelhaas, 1985; Fazzari et al., 2003; Martin, 1975]. This signature is investigated in **paper II**. However, the sources examined in **paper II** are not neutron sources.  $^{241}\text{Am}$  is a common radionuclide for gauging devices and ionizing smoke detectors.

Another impurity that is investigated in **paper II** is radioactive impurities. Since nuclides of the same element are very hard to separate there is the possibility that isotopes of the same element is present in a radioactive material, depending on mode of production. If, for example,  $^{241}\text{Am}$  is produced by separating americium from an aged plutonium stock [Comstock, 1981], there is always the possibility of presence of  $^{243}\text{Am}$  which is also formed in a nuclear reactor. The  $^{243}\text{Am}$  would in this case end up as an impurity in a  $^{241}\text{Am}$  source.





## 4 THEORY

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### 4.1 Categorization of uranium by gamma spectrometry

Uranium can be categorized by evaluating the fraction of  $^{235}\text{U}$  of the total amount of uranium. Using gamma spectrometry, this can be done by using the 185.7 keV peak from  $^{235}\text{U}$  and the 1001 keV peak from  $^{234\text{m}}\text{Pa}$  in the gamma spectrum, assuming radioactive equilibrium between  $^{234\text{m}}\text{Pa}$  and  $^{238}\text{U}$ . Four months after separation, the activity difference between  $^{234\text{m}}\text{Pa}$  and  $^{238}\text{U}$  is within the uncertainty of the gamma spectrometric measurement. Hence, radioactive equilibrium can be assumed after this period of time. The abundance of  $^{235}\text{U}$ ,  $f_{235}$ , is given by

$$f_{235} = \frac{N_{235}}{N_{235} + N_{238}} \quad (1)$$

where  $N_x$  is the number of atoms of uranium isotope  $x$ . When the enrichment of  $^{235}\text{U}$  approaches 90%, the  $^{234}\text{U}$  abundance is around 1% depending on the history of the material [Nguyen and Zsigrai, 2006]. Therefore, the amount of  $^{234}\text{U}$ ,  $N_{234}$ , is, in this case, considered to be negligible. Furthermore, the 185.7 keV peak is assumed not to be interfered by  $^{226}\text{Ra}$ . This assumption is made on the basis that there are no significant amounts of  $^{226}\text{Ra}$  in a processed anthropogenic uranium material. Moreover, the  $^{226}\text{Ra}$  originating from the background may be subtracted from the spectrum.

The radioactivity of isotope  $x$ ,  $A_x$ , evaluated from a gamma spectrometric measurement is given by

$$A_x = \frac{C_{x,\gamma}}{t \cdot I_{x,\gamma} \cdot \Psi_\gamma} \quad (2)$$

where  $C_{x,\gamma}$  and  $I_{x,\gamma}$  is the number of counts and photon emission probability of isotope  $x$  at energy  $E_\gamma$ , respectively,  $t$  is the measurement time and  $\Psi_\gamma$  is the measurement efficiency at energy  $E_\gamma$ . Using the well-known relation

$$A_x = \frac{N_x \cdot \ln(2)}{t_{1/2,x}} \quad (3)$$

where  $t_{1/2,x}$  is the half-life of isotope  $x$ , and combined with Eq. (1-2) one acquires for the abundance of  $^{235}\text{U}$

$$f_{235} = \frac{C_{235,185\text{keV}} \cdot t_{1/2,235} / I_{235,185\text{keV}} \cdot \Psi_{185\text{keV}}}{C_{235,185\text{keV}} \cdot t_{1/2,235} / I_{235,185\text{keV}} \cdot \Psi_{185\text{keV}} + C_{238,1001\text{keV}} \cdot t_{1/2,238} / I_{238,1001\text{keV}} \cdot \Psi_{1001\text{keV}}} \quad (4)$$

### 4.2 Absolute and relative efficiency calibrations

As the measurement efficiency of a gamma spectrometric system is energy dependent, the detectors need carefully performed efficiency calibrations in order to make accurate activity and isotope ratio measurements. This is normally done by using a calibration solution containing a number of radionuclides with known, certified activities and with energies covering the energy region in question. The measurement efficiency is given by rearranging Eq. 2:

$$\Psi_\gamma = \frac{C_{x,\gamma}}{t \cdot I_{x,\gamma} \cdot A_x} \quad (5)$$

Hence, the calibration spectra and the certificate information for each energy can be used to fit a response function by using an empirical equation. In this work the 5-term equation previously published by Ramebäck et al. (2010) where  $c_1, \dots, c_5$  are constants and  $E$  is the energy:

$$\Psi = e^{c_1 + c_2/E + c_3 \cdot (\ln(E))^2 + c_4 (\ln(E))^3 + c_5/E} \quad (6)$$

In special cases, where the absolute activity is unimportant such as for isotope ratio determinations, it is possible to construct a relative calibration if there is a radionuclide in the sample with a number of gamma lines covering the energy region of interest. As the activity is equal for all calibration points, Eq. 5 can be simplified and the relative measurement efficiency for a certain gamma line is then given by

$$\Psi_{\text{rel}, \gamma} = \frac{C_{x,\gamma}}{I_{x,\gamma}} \quad (7)$$

and the calculated  $\Psi_{\text{rel}, \gamma}$  can be used to fit Eq. 6 in the same manner. The advantage of using inherent calibrations is that the sample geometry is unimportant as opposed to absolute calibrations. The peaks used for the construction of an intrinsic response function for uranium abundance calculations are peaks in the low energy region for  $^{235}\text{U}$  and peaks in the high energy region for  $^{234\text{m}}\text{Pa}$ . The condition for the fitting of the function is that the  $^{234\text{m}}\text{Pa}$  peak at 258 keV is visible. This peak connects the low energy  $^{235}\text{U}$  peaks with the high energy  $^{234\text{m}}\text{Pa}$  peaks and enables a fit of a function over the whole energy region, from 144 keV to 1001 keV. For high-resolution instruments this vital peak is visible in almost all uranium spectra except possibly spectra of very highly enriched uranium. In the case of very highly enriched uranium in the high-resolution case, it would be possible to use  $^{228}\text{Th}$  daughters to establish the response function if the material contains reprocessed uranium ( $^{232}\text{U}$ ) [Ramebäck et al., 2010]. However, for low-resolution instruments this peak is not discernible and, furthermore, the low resolution reduces the number of discernible peaks for the fitting of the response function from eight to two or possibly three peaks. Hence the construction of an intrinsic response function of a low-resolution spectrum is not possible.

### 4.3 Age determination

The time that has passed since the last chemical separation is referred to as the age of a nuclear or other radioactive material. The age can be assessed by measuring the relation between the mother nuclide and its progeny. Radioactive decay can be described according to:



where  $\lambda_i$  is the decay constant for radionuclide  $X_i$ . The age of a material,  $t$ , can, in the case of two successive decays, be calculated according to

$$t = \frac{1}{\lambda_1 - \lambda_2} \cdot \ln \left( 1 - \left( 1 - \frac{\lambda_1}{\lambda_2} \right) \frac{A_2}{A_1} \right) \quad (9)$$

where  $A_i$  is the activity of radionuclide  $X_i$ .

In cases where the half-life of the daughter  $X_3$  is substantially shorter than the half-life of  $X_2$ , the  $X_3$  activity will rapidly grow into secular equilibrium and equal that of  $X_2$ . This means that  $X_3$  will decay with the half-life of  $X_2$ . In this case it is possible to use the  $A_3/A_1$  atom ratio together with the decay constant of  $X_2$  in Eq. 9 to calculate the age of the material. This may be convenient in cases where the gamma photon yield of  $X_2$  is too small to be detected with gamma spectrometry within reasonable measurement time.

## 5 EXPERIMENTAL

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### 5.1 Categorization of uranium

#### 5.1.1 Measurement setups

A number of uranium samples with varying matrix and isotopic compositions were measured using a NaI(Tl) identiFINDER® (ICx, FLIR Systems, Inc., Wilsonville, USA):

- Natural uranium (NU) as UO<sub>2</sub>
- Depleted uranium (DU) as UO<sub>2</sub>
- Low enriched uranium (LEU) with approx. 4% <sup>235</sup>U as UO<sub>2</sub>
- Natural uranium (NU) as an aqueous solution (certified reference material IRMM-184, Institute for Reference Materials and Measurements, Geel, Belgium)

For the UO<sub>2</sub> materials, data was collected for 60 s at a distance of 10 cm from the source. The aqueous reference material was measured for 600 s close to the source due to the low uranium content. The instrument gave a categorization after each measurement, and the measurement data was downloaded for off-line calculations.

#### 5.1.2 Simulations

The Monte Carlo based simulation software Virtual Gamma Spectroscopy Laboratory, VGSL, [Plenteda, 2002] was used to simulate the efficiencies for a selection of uranium materials and shielding:

- Water matrix,  $\rho = 1,0 \text{ g/cm}^3$
- UO<sub>2</sub> matrix,  $\rho = 11 \text{ g/cm}^3$
- Uranium metal matrix,  $\rho = 19 \text{ g/cm}^3$
- Water matrix with a 1 mm lead shielding,  $\rho = 11 \text{ g/cm}^3$
- Water matrix with a 5 mm steel shielding,  $\rho = 7,5 \text{ g/cm}^3$

The simulated detector was aiming to mimic the NaI(Tl) scintillation detector in the identiFINDER® instrument. The simulated data was used to fit Eq 6. The enrichment of <sup>235</sup>U was calculated for each of the spectra and response function using the peaks at 185.7 and 1001 keV for <sup>235</sup>U and <sup>238</sup>U (<sup>234m</sup>Pa), respectively.

### 5.2 Characterization of high-activity <sup>241</sup>Am sources

#### 5.2.1 Measurement setups

Five sources with the following measurement conditions were measured:

- Source 1: Nominal activity 185 GBq <sup>241</sup>Am, sealed source contained in a lead shield during gamma spectrometric measurement
- Source 2: Nominal activity 185 GBq <sup>241</sup>Am, sealed source contained in a lead shield during gamma spectrometric measurement
- Source 3: Nominal activity 3.7 GBq <sup>241</sup>Am, sealed source measured with 1.10 mm Cd shielding
- Source 4: Electroplated <sup>241</sup>Am source
- Source 5: Ionizing smoke detector

Source 1 and 2 were seemingly similar by visual inspection. Source 1-4 were measured at a 30 cm distance using a portable p-type high purity germanium gamma spectrometer (Detective-EX, EG&G

Ortec, Oak Ridge, TN, USA). Source 5, which was a smoke detector, was measured for a week using a stationary p-type coaxial HPGe detector (EG&G Ortec, Oak Ridge, TN, USA) in a lead-shielded laboratory setup.

The intrinsic efficiency response functions were established for all spectra using eight  $^{241}\text{Am}$  peaks between 208 and 802 keV and Eq. 6. Using the response functions, the activity of  $^{233}\text{Pa}$  relative to  $^{241}\text{Am}$  could be determined as well as the level of impurities compared to the  $^{241}\text{Am}$  activity.

## 6 RESULTS AND DISCUSSION

### 6.1 Categorization of uranium

The results of the instrument categorization can be found in Table 2. The instrument categorization is erroneous in all cases except for the depleted uranium sample.

Table 2 Results of the automatic instrument categorization.

Uranium type	Sample matrix	Instrument categorization
NU	UO <sub>2</sub>	DU
LEU	UO <sub>2</sub>	DU
DU	UO <sub>2</sub>	DU
NU	Aqueous	LEU

A plausible explanation to this phenomenon can be found in Figure 1. It is clear that the response varies greatly in the low energy region depending on shielding and density of the sample material. This means that if the instrument is set with a certain efficiency calibration, the evaluation of a spectrum will be erroneous if the sample characteristics differs significantly from the characteristics used to build the efficiency function. This is especially apparent in the low energy region and in the case of categorization of uranium, where a low and a high energy peak is used to calculate the abundance. A slight deviation in sample characteristics will distort the abundance calculation as the calculation of <sup>235</sup>U will be miscalculated whereas the high energy <sup>234m</sup>Pa peak is much less affected. Using a high density material response function for evaluating a spectrum of a water sample would result in overestimation of the amount of <sup>235</sup>U, hence the automatic evaluation would result in a bias, in the form of higher enrichment. This is what can be seen in Table 2 for the measurement of the aqueous solution. In the same way, if the material is denser than the response function used, the enrichment will be underestimated. This means that the accuracy of the categorization is strongly dependent on the efficiency response function used for the calculation by the instrument.

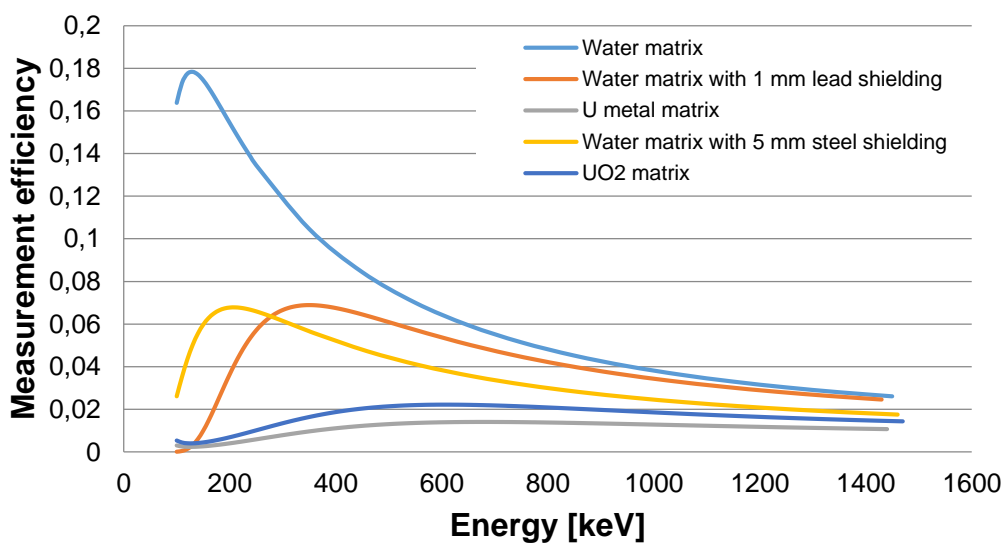


Figure 1 Simulated response functions for the investigated matrices and shieldings.

The performance criteria according to ANSI (American National Standards Institute) for hand-held instruments require that instruments should be able to identify a number of radionuclides without shielding as well as with a 5 mm steel shielding [ANSI N42.34-2006, 2007]. In fact, when evaluating the acquired spectra using the response function corresponding to a water matrix with a 5 mm steel shielding the instrument categorization results in Table 2 are reproduced. Figure 2 shows the resulting abundance of  $^{235}\text{U}$  using the response function corresponding to water with 5 mm steel shielding, different sample densities and different abundances. The orange line shows the natural abundance. This means that an instrument with a response optimized for a 5 mm steel shielding most likely will categorize uranium as DU for materials with enrichment and density corresponding to the part of the lines that are under this line. Furthermore, by using the responses in Fig. 1 it can be shown that, in order for an instrument with a setting corresponding to an inherent 5 mm steel response to show that a  $\text{UO}_2$  material is low-enriched (2% enrichment), the enrichment in the material needs to be about 15%.

To sum up, as the low-resolution instruments do not take matrix and possibility of shielding into consideration when performing the automatic categorization, there is a great risk for misinterpretation of the results. This would be avoided by using a high-resolution instrument as an intrinsic calibration easily can be constructed for each spectrum.

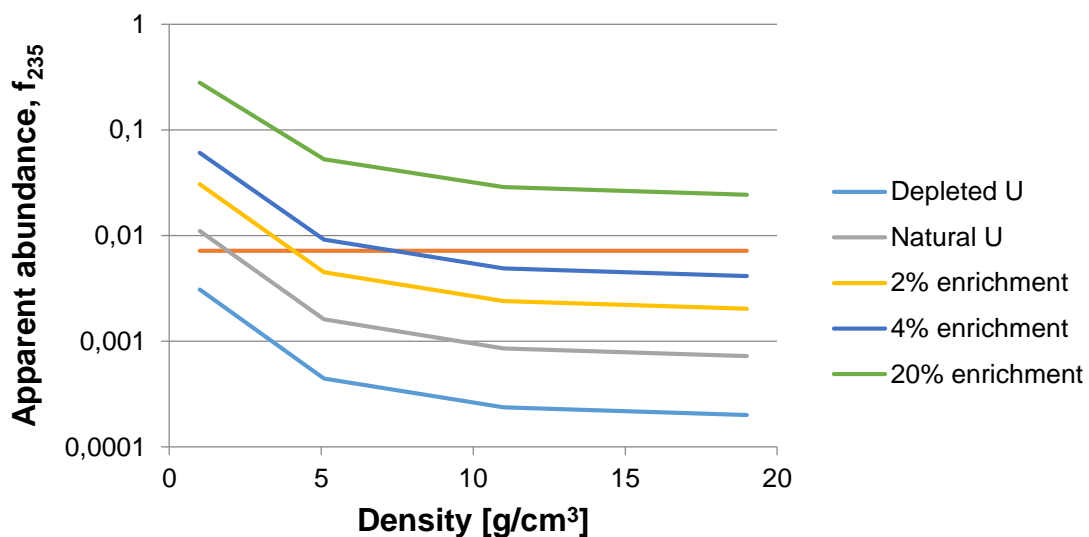


Figure 2 Apparent abundance of  $^{235}\text{U}$  using the response function, corresponding to a water sample with 5 mm steel shielding, as a function of density for different uranium containing matrices. The orange line corresponds to the abundance of natural uranium.

## 6.2 Signatures in $^{241}\text{Am}$ sources

An example of a relative efficiency response function that was established for each  $^{241}\text{Am}$  source is found in Fig. 3.

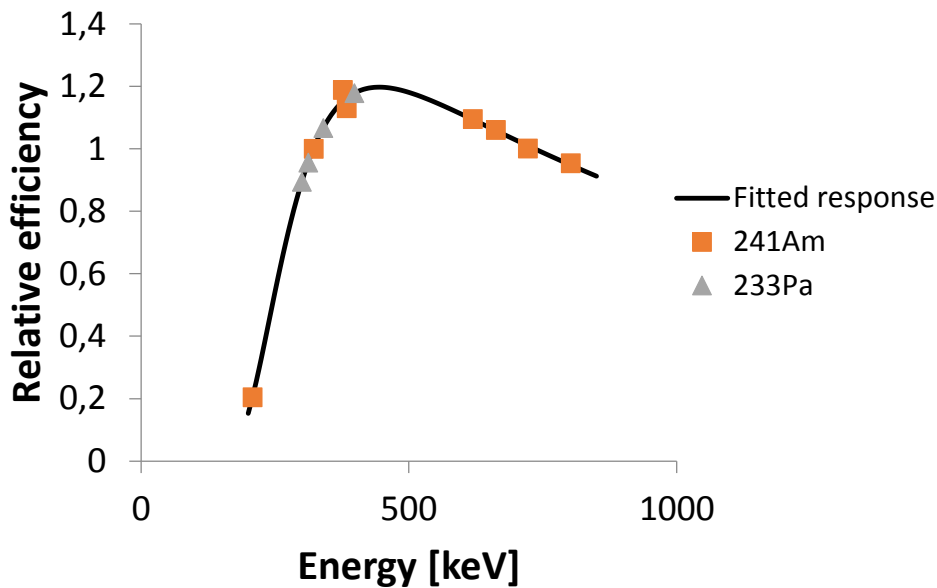


Figure 3 Efficiency response function of Source 1. The squares are the  $^{241}\text{Am}$  gamma lines used for the response function construction and the triangles are calculated responses for  $^{233}\text{Pa}$  peaks.

### 6.2.1 Age

The calculated ages of the sources are shown in Table 3. The peaks at 323 and 312 keV were used for  $^{241}\text{Am}$  and  $^{233}\text{Pa}$ , respectively. The uncertainty was calculated according to ISO: Guide to the expression of Uncertainty in Measurement (1995). The combined uncertainty consists of contributions from counting statistics, decay constants, photon emission probabilities and the response functions. The uncertainty of the response function was estimated using the jackknife procedure [Miller, 1974; Ramebäck et al., 2010]. It is also possible to calculate the age by considering the efficiency to be constant between the 323 and 312 keV peaks, thereby neglecting the measurement efficiency in the calculation. This will almost always lead to a bias. In this study, however, the difference between this method and the calculation method including the efficiency function is well within the measurement uncertainty. Hence, this alternative method can be used as a “quick and dirty”-method.

Table 3 Measured and calculated ages of the five sources. The age refers to the age at the time of measurement. The calculated separation dates are derived from the age. Uncertainties are presented with a coverage factor  $k=2$ , which corresponds to an approximate 95% confidence interval.

Source	Age [y]	$U_c$ [y] $k=2$	Separation date
Source 1	31.4	2.0	1982-01-06
Source 2	40.8	2.6	1972-07-18
Source 3	43.9	3.6	1969-07-06
Source 4	12.2	2.3	2001-06-25
Source 5	21.5	6.9	1985-07-09

The age of Source 4 was known as the material for the source was separated and electroplated at Chalmers in 2001. Hence, its age is in good agreement with the calculated age.

## 6.2.2 Impurities

### 6.2.2.1 Low-Z elements

The spectra from Source 1-3 show a number peaks that do not match peaks of  $^{241}\text{Am}$  or its progeny. Most of these peaks have an odd non-gaussian shape. The peaks seem to consist of two peaks; one part narrow and one part that is broadened at the base of the peak, see Figure 4. This peak broadening is known as Doppler broadening. Doppler broadening occurs when an atom is excited by capturing an incoming particle. This collision puts the formed atom in motion and if the atom de-excites while still in motion, the registered photons will form a broadened peak. A common example is the 511 keV annihilation peak. This peak is a result from the annihilation process when a positron, emitted during positron decay, and an electron interact and disappear and two 511 keV photons are emitted and detected. As neither the positron nor the electron is completely at rest at the annihilation, the annihilation peak is Doppler broadened since one of the photons will have a slightly larger energy than 511 keV and the other slightly smaller as a consequence of the resultant momentum [Gilmore, 2008].

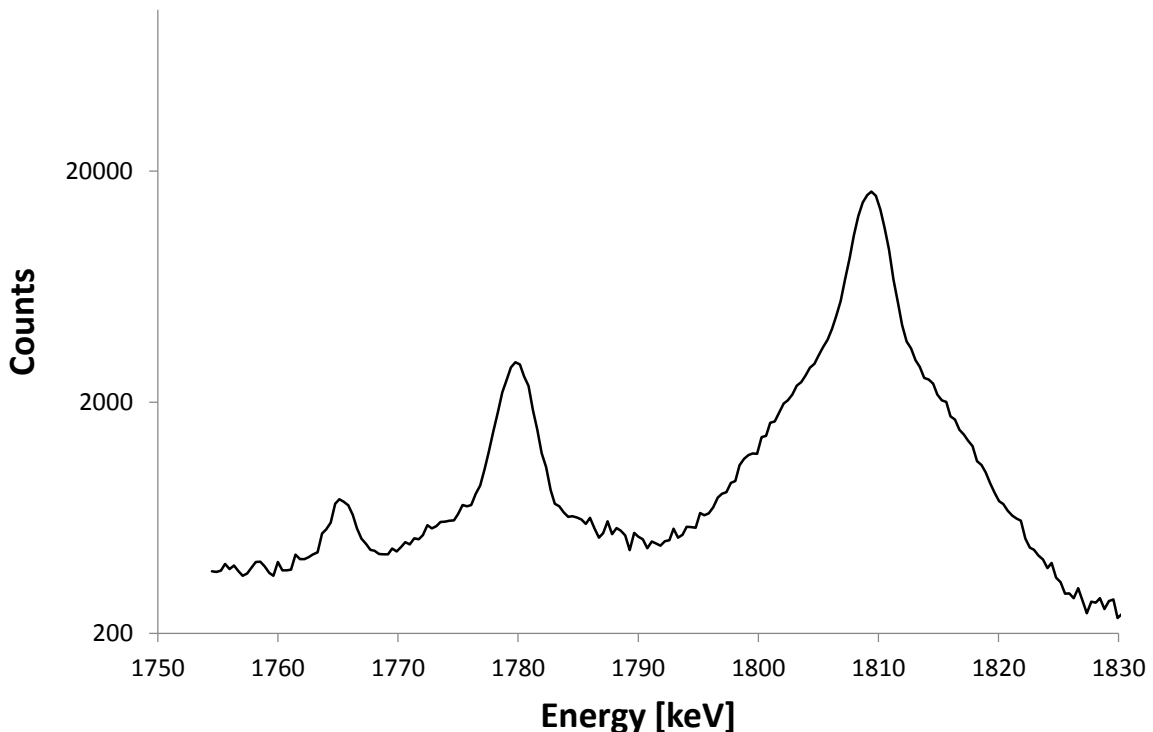


Figure 4 Example of Doppler broadened peaks.

The Doppler broadened peaks are a sign that there are low-Z elements and nuclear reactions taking place within these sources [Gehrke et al., 2003; Catz and Amiel, 1967]. The probability that a nuclear reaction will occur depends on the energy of the alpha particle. The energy must be high enough to penetrate the potential barrier caused by the Coulomb repulsion between the two positively charged nuclei. The barrier height increases with the atomic number. Hence, there is a limit to how heavy the target atom can be to be affected by a nuclear reaction. Therefore reactions are limited to elements with approx.  $Z < 20$  for most alpha emitters [Martin, 1975]. Neutron measurements confirmed the presence of neutrons in Source 1-3. Source 4-5 are sources electroplated on steel discs, hence there are no low-Z elements present. Accordingly, the spectra do not show any broadened peaks due to the Doppler Effect. The deceleration time of  $^{26}\text{Mg}$  and  $^{29}\text{Si}$  nuclei in an  $^{241}\text{Am}$  source was calculated to vary from approx. 400 fs to 500 fs for  $^{26}\text{Mg}$  and from approx. 350 fs to 450 fs for  $^{29}\text{Si}$  [Vesterlund et al.,



2015]. This means that if the de-excitation half-life of an excited atom is shorter than about 500 fs, the decay will add to the broadened part of the peak. Suggested reactions and related gamma lines are presented in Figure 5. Some of the lines could be attributed to  $^{23}\text{Na}$ , see Figure 5. It is suggested that the sources consist of a core made from fused glass which would explain the presence of sodium. Other spectrum peaks correspond to reactions on  $^{26}\text{Mg}$ . Whether the magnesium is present in the encapsulation or has been built up from the  $^{23}\text{Na}(\alpha, p)^{26}\text{Mg}$  reaction is difficult to tell. One way of knowing is to look at the presence of peaks of other magnesium isotopes and see if the magnesium is of natural composition, but unfortunately these peaks are not exclusive to magnesium.

Another possible signature is to look at the relation between sodium and  $^{241}\text{Am}$  in the sample sources. The 440 keV peak originating from the  $^{23}\text{Na}(\alpha, \alpha')^{23}\text{Na}$  reaction could be used for this purpose. To estimate the sodium impurity level the ratio between the relative activities at 440 keV and 619 keV was calculated according to

$$R_{\text{Na}/\text{Am}} = \frac{S_{440}/\Psi_{440}}{S_{619}/\Psi_{619}} \quad (10)$$

where  $S_E$  and  $\Psi_E$  is the peak area and measurement efficiency at energy E keV, respectively. The 440 keV sodium peak is interfered by two weak  $^{241}\text{Am}$  gamma lines. These contributions were subtracted using the peak area at 619 keV.

*Table 4 Ratios of measurement efficiency corrected peak areas at 440 and 619 keV. Uncertainties are presented with a coverage factor  $k=2$ , which corresponds to an approximate 95% confidence interval.*

	$R_{\text{Na}/\text{Am}}$	$U_c, k=2$
Source 1	0.1854	0.0098
Source 2	0.260	0.014
Source 3	0.286	0.056

The results of the calculations can be seen in Table 4. One question that needs to be raised is the relation between amount of sodium and age. Will the sodium in the sources deplete due to the nuclear reactions such that the measured ratio changes with time or is the depletion so small that the change with time is insignificant within a reasonable timeframe and thus can be considered constant? This has, however, not been a subject in this work.

Source 1 and Source 2 have the same nominal activity, 185 GBq. However, the results in Table 4 show that Source 1 and 2 have significantly different Na/Am-ratios. This could imply that the amount of sodium is higher in Source 2 than in Source 1. Source 3 has a higher ratio than Source 1 and 2. The reason for this could be that Source 1 and 2 have an activity and hence an alpha flux which is 50 times higher than the activity of Source 3. This could lead to lower sodium depletion in Source 3 and hence a higher ratio. Another reason may be that the initial sodium concentration was higher in Source 3 than in the other sources. The difference between the ratio of Source 2 and 3 are not statistically significant, however.

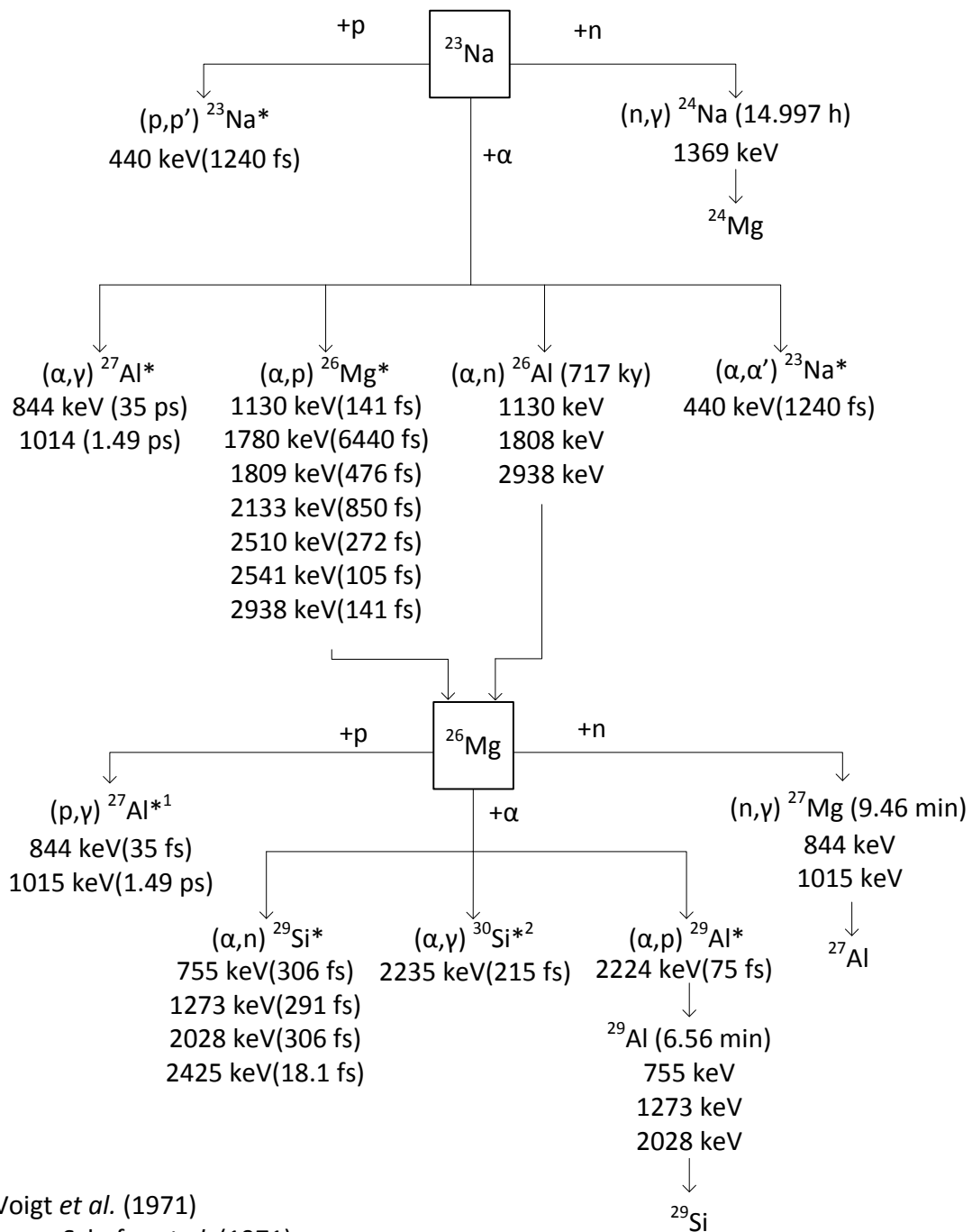


Figure 5 Identified gamma lines from radionuclides other than  $^{241}\text{Am}$  and its progeny, and suggested reactions based on impurities. The radionuclides with an asterisk are emitting gamma rays due to de-excitation. The reference to the gamma lines and corresponding half-lives are taken from the Nudat 2.6 database (2014) unless otherwise stated.

### 6.2.2.2 $^{243}\text{Am}$

In two of the spectra, Source 1 and 3,  $^{239}\text{Np}$  could be identified. A comparison between a  $^{239}\text{Np}$  containing and a non- $^{239}\text{Np}$  containing spectrum from Source 1 and 2, respectively, can be seen in Figure 6.

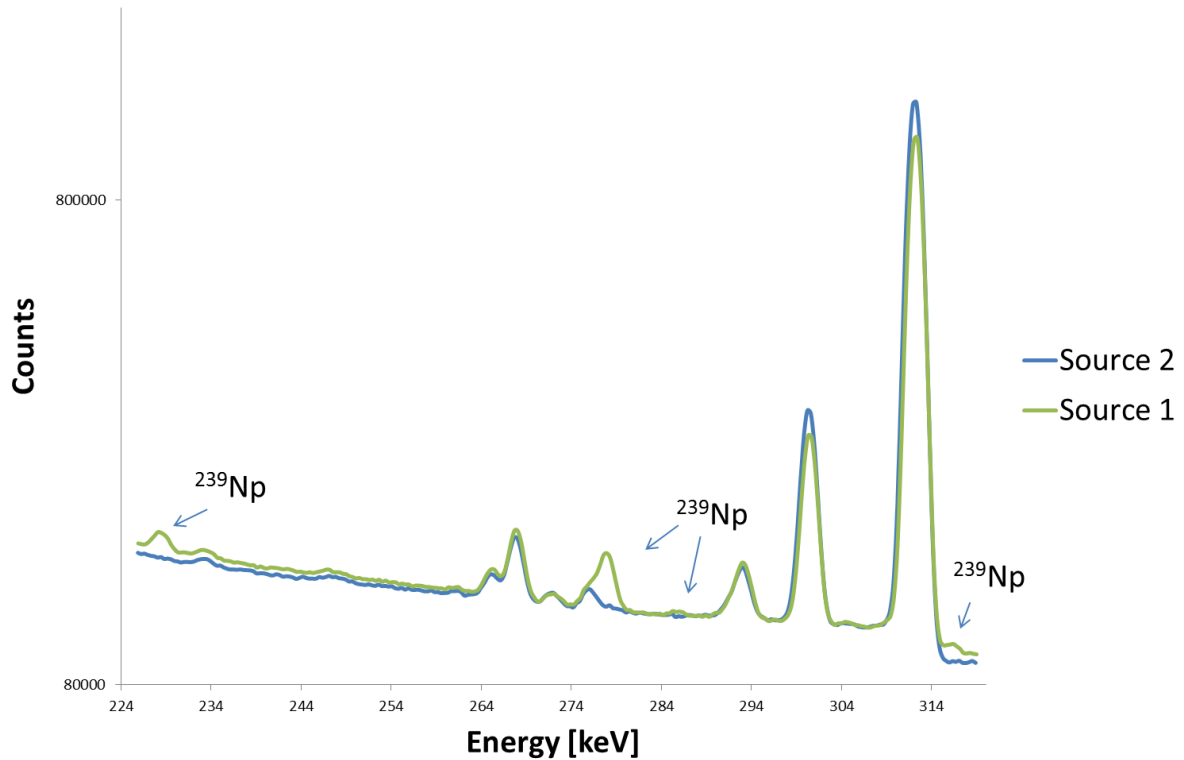


Figure 6 Comparison of Source 1 and Source 2. Source 1 show  $^{239}\text{Np}$  peaks as opposed to Source 2.

The presence of  $^{239}\text{Np}$  could imply that these sources contain  $^{243}\text{Am}$  as an impurity as  $^{239}\text{Np}$  is the daughter of  $^{243}\text{Am}$ . The photon emission probabilities for  $^{243}\text{Am}$  gamma lines are, however, too weak to be visible in the spectra from these measurements. Since the half-life of  $^{239}\text{Np}$  is 2.356 days [DDEP, 2015]  $^{239}\text{Np}$  is in secular equilibrium with  $^{243}\text{Am}$ . It is therefore possible to calculate the  $^{243}\text{Am}/^{241}\text{Am}$  ratio. The  $^{243}\text{Am}/^{241}\text{Am}$  ratio is  $1.444(48)\cdot 10^{-6}$  and  $2.09(11)\cdot 10^{-7}$  for Source 1 and Source 3, respectively, where the numbers in parentheses, which are the numerical values of the combined uncertainty, refer to the corresponding last digits of the quoted results.



## 7 CONCLUSIONS

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The work on categorization of uranium shows that it is difficult to use low-resolution instruments to automatically identify the uranium category as the identification algorithm does not take the sample matrix and the possibility of a shielded sample into consideration. Even though a number of methods have been proposed in the literature over the years, the categorization ability of the commercially available instruments seem to remain uncertain. The results implies that if using a low-resolution instrument for categorization, the certainty of the results could improve if the instruments were equipped with a number of efficiency response functions for different sample conditions to make it possible to test different response functions. In this way the instrument could provide the user with a range of categories, making it possible to narrow down the number of measures following uranium detection in for example a border control. Another possible option, also depending on the knowledge of the sample conditions, could be to take the data offline and perform the data evaluation manually. It would then, however, be more convenient to use a high-resolution instrument at once, since these instruments have better capacity to perform the right category evaluation.

The work on strong Am-sources shows that information found inherent in the sources have good potential to be used as signatures for sources containing alpha-emitting radionuclides and some stable material within the source, either as impurities or as support material. The work shows that all investigated sources could be differentiated between by using only one gamma spectrum acquired from each source. From each gamma spectrum the age of the sources could be calculated. In a couple of the spectra, traces of  $^{243}\text{Am}$  could be seen and the ratio between the  $^{243}\text{Am}$  and  $^{241}\text{Am}$  differed in these spectra. This information can also be used as a signature for a specific source. A third signature was the presence of low-Z elements in the source which together with the alpha-particles generate nuclear reactions within the sources which are seen as Doppler broadened peaks in the spectra. Varying composition of stable elements in sources would provide different nuclear reactions and hence different peaks in the gamma spectra. All this information could be used to populate a national nuclear forensic library which would make it possible to, if not single out a specific source, at least narrow down the number of candidates in the case of a source out of regulatory control.



## 8 FUTURE WORK

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The continuing work will move on from signatures found by gamma spectrometry measurements to signatures in uranium, measurable by ICP-MS. ICP-MS provides the possibility to measure low or ultra-low concentrations of stable isotopes or long-lived radioactive isotopes. Stable elements are present as either impurities or as additives in uranium. To be able to measure these low concentrations in uranium-bearing material it is necessary to remove the uranium to avoid matrix effects in the plasma and unnecessary instrument contamination.

One group of elements that is interesting as signatures are common transition metals that are used throughout the fuel production process as alloy metals in tools in different production stages or as production chemicals. This signature could provide information about the production processes the investigated uranium material has undergone. Another group of elements that are put forward as a promising signature for uranium is the lanthanides. These elements have similar chemical properties and are therefore considered to behave similarly throughout the uranium fuel production process. Hence, it is suggested that the ratio between the lanthanides, the so called lanthanide pattern, will remain undisturbed and therefore is a good signature for the geographical origin of the uranium material as well as the type of uranium deposit. A problem with lanthanide measurements with ICP-MS are the interferences caused by isobars and polyatomic species such as oxides. The lanthanides are prone to oxide formation in the plasma which causes overestimation of foremost the heavier lanthanides [Vesterlund et al, 2014]. Therefore the future work will focus on developing separation and measurement methods for accurate measurements of stable isotopes using ICP-MS.





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