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DIPLOMARBEIT

Hardware and ID Software Test to identify selected Isotopes under preset Conditions

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Abstract

Illicit trafficking of radioactive material is a serious problem in context with the arising possibilities of international terrorism. The mission of the International Atomic Energy Agency (IAEA) is to enhance the ability to detect nuclear materials in the field (e.g. border crossings). Small devices called “Handheld RID’s” (Radionuclide Identifying Device) which are lightweight, mobile and capable of identifying radiating materials are developed. The IAEA is in tight contact with manufacturing companies to assess the performance and to systematically improve the devices. Those efforts have yield fruit in the meantime but cannot be incorporated to the full extent into the state of 4 years ago described in this work.

This work will examine the ability of some RID’s to identify certain isotopes under preset conditions and the software used to analyse the recorded spectra.

The first goal of this work is to discover the detection limits of the detectors (ICx IdentiFinder, Aspect MKC A03) as well as the ID software (IdentPro) and analyse those limits to arrive at conclusions for further development of both components. Another part concentrates on masking threatening isotopes (SNM) with, for example medical isotopes and the ability of software algorithms to detect a masked isotope.

The results show that there are noticeable differences in the quality of detection – comparing hardware and software as well as the software products among each other. Both detecting devices as well as the software algorithms have their advantages as well as their failings. The quality of the results depends strongly on the type of isotope or the isotope-mixture.

Kurzfassung

Illegaler Handel mit radioaktivem Material ist ein ernstes Problem im Kontext der aufkeimenden Möglichkeiten des internationalen Terrorismus. Eine Hauptaufgabe der Internationalen Atomenergie Behörde (IAEA) ist die Verbesserung der Möglichkeiten zum Nachweis von radioaktivem Material in der praktischen Anwendung (z.B. an Grenzübergängen). Kleine, leichte und tragbare Geräte, bekannt als „Handheld RID’s“ (Radionuclide Identifying Device), die außerdem in der Lage sind radioaktive Materialien zu identifizieren werden entwickelt. Die IAEA ist in Kontakt mit führenden Geräteherstellern um a) die Performance einzuschätzen, und b) die Geräte gezielt zu verbessern. Diese Bemühungen haben inzwischen Früchte getragen, die im hier dargestellten Stand von vor 4 Jahren nicht vollständig berücksichtigt werden konnten.

Diese Arbeit beleuchtet die Leistungsfähigkeit von einigen RID’s in Bezug auf die Erkennung von radioaktiven Substanzen unter klar definierten Bedingungen.

Das erste Ziel ist die Definition von Erkennungsgrenzen für Detektoren (ICx Identifinder, Aspect MKC A03) und ID Software Produkte (IdentPro) und wie diese Ergebnisse genutzt werden können um die Entwicklung voranzutreiben. Ein anderer Teil der Arbeit beschäftigt sich mit dem Problem der Maskierung von gefährlichen Isotopen (SNM) mit medizinischen Isotopen und der Fähigkeit der Software Algorithmen das maskierte Isotop klar zu identifizieren.

Die Ergebnisse zeigen erhebliche Unterschiede in der Qualität der Erkennung – sowohl im Vergleich zwischen Hard- und Software als auch im direkten Vergleich der Software-Produkte. Beide Detektoren und die Software Algorithmen haben ihre Stärken, aber genauso signifikante Schwächen. Die Qualität der Ergebnisse hängt maßgeblich vom Isotop bzw. vom Isotopengemisch ab.

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1. Introduction

“US Nuclear Weapons Site in Europe Breached”. That was a headline of a news-article found on the website of the Federation of American Scientists (Hans Kristensen, 2010). The story tells about some peace-activists who were able to enter a US military base in Belgium rather trouble-free. They walked around uninterruptedly for about an hour before some security guard asked what they were doing next to where the U.S. Air Force currently stores about 10-20 nuclear bombs...

One should think that nowadays, where terroristic acts are on everyday's headlines the awareness of the threat of nuclear material should be in everybody's mind.

At least in the mind of our public authorities.

Even in central Europe the fear of “dirty bombs”¹ and smuggling of nuclear material or devices increases.

One of the main pillars of the International Atomic Energy Agency (IAEA²) is to prevent the further spread of nuclear weapons. Illicit Trafficking of nuclear or radioactive material is a serious problem in context with the arising possibilities of international terrorism.

The IAEA in close collaboration with local governments tries to prevent terrorist attacks and smuggling by very strict surveillance e.g. of borders, airports and main public events like for example the soccer world cup.

The intent of all those efforts is, to provide a certain security-level for the people without letting them know that they are scanned for radioactive isotopes at the airport or while watching a soccer game in a stadium.

¹ http://en.wikipedia.org/wiki/Dirty_bomb, March 6th 2010

² <http://www.iaea.org/OurWork/index.html>, March 6th 2010

The innocent citizen should not even be bothered by the surveillance and therefore one is in the need of devices, which can distinguish between a person who smuggles uranium and a person who just had a medical treatment with radioactive iodide.

The mentioned devices are so called "Radionuclide Identifying Devices", in short "RIDs".

In this work the features and application range of handheld RIDs (small, portable devices), their physical background, and ability to identify certain isotopes under preset, sometimes complicated conditions will be discussed. An important part of this work is the software, which is used to examine spectra recorded by the devices. It is important to see, which vantages and disadvantages of both, devices and software are standing out.

In another part of this work the attention is concentrated on masking threatening isotopes with, for example medical isotopes and the impact of this issue on the further development and the requirements of new RIDs and the recognition software.

A detailed discussion of the advantages, disadvantages and future possibilities of the treated devices as well as of the software used to process the recorded data will conclude this work.

An outlook to the future of border monitoring and the idea of a cross-linked incident database will give something to think about.

2. Tasks and Motivation

This section will give a short input about what RIDs are. What are the differences to other groups of devices which measure only quantitative characteristics of radiating materials? Why is it important to discover which isotopes are actually causing the radiation measured?

There will be an overview of existing designs, types and their range of application. A few practical examples will explain the work with such devices and give an idea of what the devices should be able to accomplish.

2.1. What are RIDs?

When monitoring people, trucks or luggage it is of great interest if there are any hidden, radiating materials present. In order to detect those materials, the emitted radiation has to reach the measuring devices and therefore penetrate any surrounding materials or containers. Since the radiating materials are usually not passed by the detector on a silver tablet, they are typically surrounded either by a suitcase or a backpack, a container or a human body.

That is why there is almost no possibility to detect alpha radiation, low energy beta radiation and low energy gamma radiation, which can easily be shielded. In case of alpha radiation and low energy beta radiation a few layers of paper or the human body would be sufficient, whilst low energy gamma radiation could be shielded by a steel container. It is therefore obvious that it doesn't make sense to put any effort in detecting alpha or beta radiation.

Another type of radiation which can be detected is neutron radiation. Due to the fact that sources for neutron radiation do not exist as naturally occurring radioactive materials, neutron radiation can be used as an indicator for nuclear material.

That is why detection devices focus on the ability to recognise nuclear gamma sources and neutron radiation.

There are many devices which measure quantitative characteristics of radiating materials such as a Geiger counter or dosimeters. They give an indication of the intensity of the examined radiation and tell us whether the radiation is dangerous or not but the operator cannot make a statement on the type and characteristics of the radiating source.

Unfortunately this information can be very important, for example if people on an airport are monitored. The aim is to have the possibility to say whether a person is smuggling radioactive materials or just had a medical treatment with radioactive sources like Technetium (Tc-99) or Iodide (I-131).

To find out which isotopes cause the detected radiation the devices avail themselves of gamma-spectroscopy.

Gamma radiation is an electromagnetic radiation and is often produced alongside other forms of radiation such as alpha or beta radiation. When a nucleus emits an alpha or beta-particle, the residual daughter nucleus is sometimes left in an excited state. The excited nucleus can then relax to the ground state by emitting one or more gamma quanta of characteristic energy.

The gamma-emission spectrum is therefore used to identify the radiating isotopes.

Radionuclide Identifying Devices (RIDs) or sometimes Radio Isotope Identifying Devices (RIIDs) are a class of devices which are able to identify the source of gamma radiation by recognising the characteristic gamma-spectrum.

How the devices manage to identify the characteristic pattern of a certain isotope is described later on.

Depending on the field of application there exist various types of devices.

Generally, the International Atomic Energy Agency (IAEA) distinguishes between 3 types of instruments³:

³ (Radiation Safety Section - International Atomic Energy Agency, 2002), p. 6

1. Pocket type instruments, which are very small and lightweight (PDA-size). They are usually used to inform the operator of a present activity and of the actual radiation level. Actually some devices have a “search” - mode to look for hidden nuclear material. The newer generations of this instrument-class are so called “Spectroscopic Personal Radiation Detectors” (SPRD) which are able to identify radioactive isotopes and therefore fit into the group of RIDs. Due to the compact size, the sensitivity and accuracy of those devices is not too good.
2. Hand-Held instruments are of bigger size and can usually not be worn covertly. They feature bigger detection units and therefore higher sensitivity and higher performance at dose rate measurements and isotope identification. Nowadays most devices of this class are RIDs. This work will focus on this class of devices.
3. Fixed installed, automatic systems are used to monitor and observe people, luggage and vehicles. Such systems are installed at airports, seaports and at border crossings for road and railways. They manage to cope with a high flow of vehicles or persons. To provide the needed sensitivity the used detectors are of large volume. Most of these devices are only used to discover nuclear material. Only few systems are also able to identify radioactive isotopes. They are used for example at airports to monitor passengers.

2.2. What are RIDs used for?

As mentioned above RIDs are able to identify radioactive isotopes rather than just give information of whether there is radiating material present and what is the actual dose rate.

This chapter will give a review of the various applications of RIDs and describe the workflow at border crossings.

Some of the possible fields of application are:

- Search and Analysis of Special Nuclear Material (SNM) and other Norm or Industrial radionuclides
- Surveillance of major public events like Soccer World Cup or Olympic Games

- Safeguard Activities (e.g. Safeguard Inspectors for the IAEA)
- Customs
- Monitoring of border crossings
- Surveillance of airports and harbours
- Atomic Safety and Radiation Protection at nuclear facilities or hospitals
- Police and Fire Brigade
- Waste monitoring and analysis
- Search for radioactive contaminated ammunition (depleted uranium)
- Emergency Nuclear Events
- Contamination Monitoring

2.2.1. Surveillance of major public events

To prevent visitors from smuggling radioactive material onto the events venue, one uses pedestrian monitors in combination with hand held RIDs. The pedestrians have to pass a bottleneck to enter the location. At this point they are scanned, by a hidden Pedestrian monitor, which is able to decide whether people passing by wear medical isotopes or some kind of dangerous, nuclear material. Officers can easily separate conspicuous persons and examine them using the RIDs.

Another way of keeping the venue of such events under surveillance are helicopters equipped with huge detector-units, which circle above the location and scan for nuclear material.

Recent developments for this purpose are so called backpack systems. These are more or less standard type backpacks, which contain for example a gamma sensitive detector and a device for neutron detection together with all the necessary read-out electronics, a multi channel analyser (MCA), a sub-notebook to record and store the spectra and various features for connectivity such as Bluetooth or a GPS-receiver. Some advantages of such systems are that they are lot cheaper then helicopters and that they are not as eye-catching. The officer wearing the backpack is furthermore able to respond immediately to occurring complications.

2.2.2. Safeguard activities

One of the three main areas of work of the IAEA is "Safeguards and Verification". Inspectors work to verify that safeguarded nuclear material and

activities are not used for military purposes. Most of these activities do not reach the public, but in the recent past everybody was aware of the work of IAEAs inspectors in Iraq, where Saddam Hussein reportedly hoarded nuclear weapons, which - of course - couldn't be found up to the present.

The inspectors use various devices to search for and identify nuclear material.

2.2.3. Police and Fire Brigade

In Austria it is not standard, that task forces like police or fire brigade are equipped with devices for the detection and identification of radioactive materials. In case of an emergency case, where radiating materials are involved, the task forces have to contact institutions like the Institute of Atomic and Subatomic Physics⁴ (former Vienna Atomic Institute of the Austrian Universities) to borrow hand held devices for nuclide identification.

In other countries like the USA, police officers always carry small devices (Size of a PDA or pager) called PRDs (Personal Radiation Detectors) with them, which are used to inform the officer of deviating or risen radiation levels. Those small devices are normally not able to identify the radiating source, so there have to be RIDs available at the police station.

A more sophisticated coming up class of devices, SPRDs (Spectroscopic Personal Radiation Detector) are also capable of isotope identification, while the size is still not larger than a PDA. A good example for a SPRD is the "Interceptor", developed by Target Instruments for Thermo Electron Corporation⁵.

2.2.4. Customs and Border Monitoring

Further common applications of RIDs are customs controls and security at border crossings, road as well as train. The technology is also utilised by customs authorities at harbours (container) and airports.

It is almost impossible to check every vehicle, every train and every container for illicit transportation of nuclear material. The solution is an automated system of so called portal monitors. Portal monitors are fixed installed devices, where vehicles can pass through. The systems are able to detect even small amounts of radiation and cause an alarm in case of inconsistencies. The vehicle is stopped and customs officers examine the vehicle with hand held RIDs to find out if there really is any radiating source and whether the source is illegal or dangerous. It is

⁴ <http://www.ati.ac.at>, March 13th 2010

⁵ <http://www.thermo.com/com/cda/product/detail/1,1055,10122134,00.html>, March 13th '10

important to distinguish between natural occurring radioactivity, medical isotopes and material which is illegal. For example fertiliser contains Potassium, which consists of 0,012% K40 - a natural radioactive isotope of Potassium. The portal monitors at borders are in general not able to do isotope identification and therefore they will trigger an alarm when a truck with 15 tons of fertiliser passes the device. Customs officers will find out rather quickly that the detected radiation is natural and does not pose a threat.

Specifications for hand held RIDs say that they have to be easy to use by non-professional personnel like customs officers or policemen. Non-professional is meant in terms of scientific radiation detection and gamma spectroscopy.

Most countries have specific authorities to assist customs officers in cases of the detection of nuclear material. Those authorities also collect the data of incidents occurring at their borders. The IAEA is eager to gain this data to provide it to all member states in order to trace smuggled material and build up some kind of knowledge base.

It is also the job of the IAEA to assist the member states with purchasing the equipment for radiation detection and to train the people who have to work with those devices.

Illicit trafficking is an increasing problem and therefore the work of customs authorities around the world is getting more and more important.

It is of course no problem to continue this list of possible applications of RIDs and in the course of this work there will be some room to discuss some of those and explain which devices fit best for different tasks.

The next section is about the International Atomic Energy Agency, the mission, the work and common tasks the IAEA has to fulfil.

3. Physical background

In order to talk about the detection of radioactive isotopes or nuclear material, one has to understand how the detection works and on what physical principles it bases. This chapter gives a brief overview on the radioactive decay, the detection of emitted gamma radiation and the various algorithms used by software for identifying isotopes based on a gamma spectrum. Some types of radiation detection devices are being introduced as well as necessary electronics to record gamma spectra.

3.1. Radioactive decay & gamma emission

The radioactive decay is a stochastic process in which an unstable atomic nucleus transforms spontaneously under the emission of ionizing radiation (subatomic particles and/or electromagnetic waves). The emission of either radiation corresponds to a loss of energy, which – in case of gamma radiation – is specific for the decaying nucleus.

Because of this and the fact that gamma radiation occurs in a large number of nuclear transformations, it is evident, that the examination of gamma radiation is of particular importance to the field of isotope identification.

3.1.1. Radioactive decay

Radioactivity is the property of atomic nuclei with an unstable nuclide configuration to perform a spontaneous transformation under the emission of energy.

While stable nuclides exist unchanged for an indefinite period of time, the unstable nuclides are radioactive and they decay.

A nuclide is unstable if

1. There is another nuclide configuration with a lower total energy

2. There is a process which can reach this configuration while obeying the conservation rules

The unstable nucleus always wants to reach a state of greater entropy. The system will move towards the ground state, it will spontaneously stabilize to a less-excited system. The transformation alters the structure of the nucleus and results in the emission of either photons or a high-velocity particle (such as an alpha particle, an electron, a neutron,...).

The mass of the daughter nucleus (product, after the decay) is smaller than the mass of the primary nuclide (parent nucleus) and the mass difference between the two states can be quantified as the energy released during the decay. This can be expressed using Einstein's formula $\Delta E = \Delta m \times c^2$, where ΔE [J] is the released energy, Δm [kg] the mass difference and c [ms^{-1}] the speed of light in vacuum.

Since the radioactive decay is a stochastic process, it is only possible to give information on a large amount of atomic nuclei but there is no way to predict the exact moment of the decay of a specific nucleus.

In a given system of N unstable atomic nuclei, the reduction/decay of nuclei per time unit is proportional to the number of nuclei.

$$\frac{dN(t)}{dt} = -\lambda N(t) \quad (3-1)$$

The proportional constant λ [s^{-1}] is called the decay constant and is characteristic for each isotope. It is also often referred to as transition probability.

To get the number of nuclei at a given time, the formula above is being transformed to the following, describing an exponential decay.

$$N(t) = N_0 \cdot e^{-\lambda t} = N_0 \cdot e^{-\frac{1}{\tau}t} \quad (3-2)$$

N_0 describes the number of nuclei at the time $t=0$.

The decay time $\tau = 1/\lambda$ describes the mean life time of the nuclei.

The time during which half of the nuclei have decayed is called half life period $T_{1/2}$ [s], which is defined as

$$T_{\frac{1}{2}} = \frac{\ln 2}{\lambda} \quad (3-3)$$

The number of decays per given time is called activity. The SI unit of the activity is the Becquerel (Bq [s^{-1}]), which is defined as one transformation / decay per second. Since the activity $A(t)$ of the sample changes over time, the date of measurement has to be declared together with the information on the activity.

$$A(t) = -\frac{dN(t)}{dt} = \lambda N = \lambda N_0 \cdot e^{-\lambda t} = A_0 \cdot e^{-\lambda t} \quad (3-4)$$

Resulting radiations from a nuclear decay have been classified according to their ability to pass through matter.

- α particles (He^{2+})
- β particles (electrons e^- or positrons e^+)
- γ rays (electromagnetic radiation)

3.1.2. Emission of gamma radiation

Gamma rays are often produced alongside other forms of radiation such as alpha or beta radiation. When a nucleus emits an alpha or beta particle, the daughter nucleus is sometimes left in an excited state. It can then de-excite to a lower energy state by emitting a single or as well series of gamma rays, in much the same way that an atomic electron can jump to a lower energy state by emitting light (infrared, visible, or ultraviolet). That is why gamma radiation is actually not really a decay mode, rather than a de-excitation of an excited daughter nucleus resulting from a β -decay⁶. The daughter nucleus in its ground state might then decay further or be stable.

Gamma rays are the most energetic form of electromagnetic radiation. Even though their energy is in some cases much the same as the energy of x-rays, their origin varies to the origin of x-rays. While gamma rays are emitted by the nucleus or from other particle decays or annihilation events, x-rays are emitted by electrons outside the nucleus. High energetic radiation corresponds to very short wavelengths as the following relationship explains:

$$E = h \cdot \nu = \frac{c}{\lambda} \quad (3-5)$$

⁶ (Knoll, 1979)

Where $h = 4,136 \cdot 10^{-15} \text{ eVs}$ is Planck's constant, ν is the frequency and λ the wavelength of the emitted gamma quantum.

It is obvious that the emitted gamma ray can only carry a discrete amount of energy (limited to multiples of h) which is corresponding to the origin process of the radiation.

In the following example Co-60 transforms under a β^- -decay to an excited state of the Ni-60 nucleus which relaxes under the emission of two photons to the ground state of Ni-60. The resulting gamma-radiation with the energies $\gamma_1 = 1173\text{keV}$ and $\gamma_2 = 1332\text{keV}$ is specific for the decay of Co-60 and can therefore be used to identify this radioactive nuclide. Those energies show up as discrete lines in the energy-spectrum and each of them arises from a transition between two energy states of the excited nucleus (in this case Ni-60).

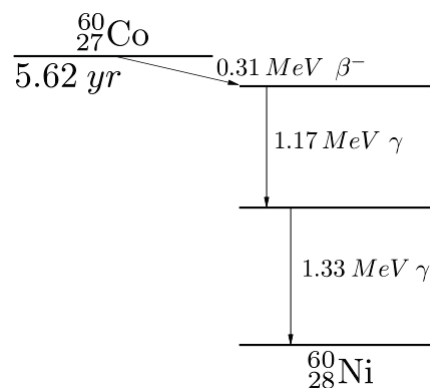


Fig. 3-1 Decay scheme of cobalt-60

3.2. Interaction of gamma radiation with matter

The detection of gamma rays depends on their interaction with matter. The energy of the gammas can be transferred to the electrons within the detector material through different processes. These processes are photoelectric absorption, Compton scattering and pair production. In semiconductor detectors, all of the three interaction processes generate free electrons when the gamma rays pass through matter. These electrons slow down as they create electron-ion or electron-hole pairs. One can use these pairs to determine the energy of the passing photon by measuring the quantity of charge created since

the number of electron-hole pairs is proportional to the energy of the electrons produced by the primary interaction.

Photoelectric interactions are dominant at low energy, Compton scattering in the mid-energy range and pair production only occurs above about twice the rest energy of electrons, 1022 keV. The following paragraphs will outline the individual interaction processes and the overall attenuation of a gamma ray in matter.

Although a large number of possible interaction mechanisms are known for gamma rays in matter, only three major types play an important role in radiation measurements: photoelectric absorption, Compton scattering, and pair production. All of these processes lead to the partial or complete transfer of the gamma ray photon energy to electron energy. They result in sudden and abrupt changes in the gamma ray photon history, in that the photon either disappears entirely or is scattered through a large average angle. This behaviour is in marked contrast to the charged particles discussed earlier in this chapter which show down gradually through continuous, simultaneous interactions with many absorber atoms. The fundamentals of the gamma ray interaction mechanisms are introduced here, but are again reviewed at the beginning of Chapter 10 in the context of their influence on the response of gamma ray detectors.

3.2.1. Photoelectric absorption

In the photoelectric absorption process, an incoming gamma ray photon undergoes an interaction with an absorber atom in which the photon completely disappears. In its place, an energetic photoelectron is ejected by the atom from one of its bound shells. The interaction is with the atom as a whole, and cannot take place with free electrons. For gamma rays of sufficient energy, the most probable origin of the photoelectron is the most tightly bound or K shell of the atom. The photoelectron appears with an energy given by

$$E_e = h\nu - E_b \quad (3-6)$$

where E_b represents the binding energy of the photoelectron in its original shell. For gamma ray energies of more than a few hundred keV, the photoelectron carries off the majority of the original photon energy.

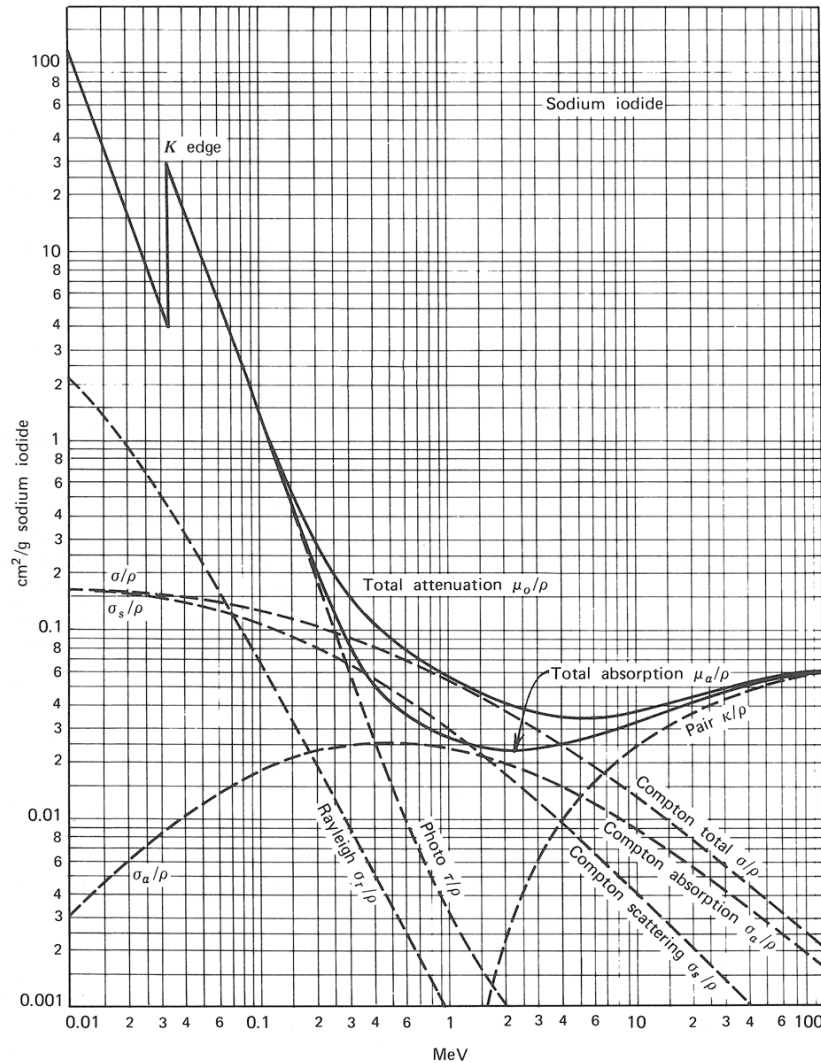


Fig. 3-2 Energy dependence of gamma ray interaction processes in sodium iodide⁷

In addition to the photoelectron, the interaction also creates an ionized absorber atom with a vacancy in one of its bound shells. This vacancy is quickly filled through capture of a free electron from the medium and/or rearrangement of electrons from other shells of the atom. Therefore, one or more characteristic X-ray photons may also be generated. Although in most cases these X-rays are reabsorbed close to the original site through photoelectric absorption involving less tightly bound shells, their migration and possible escape from radiation detectors can influence their response. In some fraction of the cases, the

⁷ (Evans, 1955)

emission of an Auger electron may substitute for the characteristic X-ray in carrying away the atomic excitation energy.

The photoelectric process is the predominant mode of interaction for gamma rays (or X-rays) of relatively low energy. The process is also enhanced for absorber materials of high atomic number Z . Many detectors used for gamma ray spectroscopy are chosen from high- Z constituents for this reason.

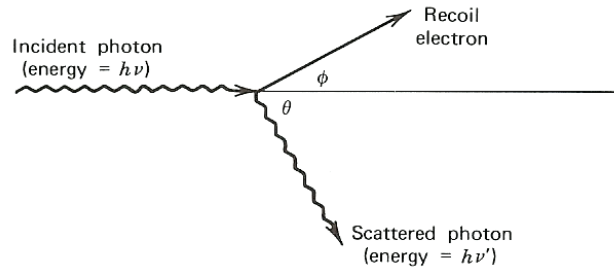
A plot of the photoelectric absorption cross section for a popular gamma ray detection material, sodium iodide, is shown in Fig. 3-2. In the low-energy region, discontinuities in the curve or "absorption edges" appear at gamma ray energies which correspond to the binding energies of electrons in the various shells of the absorber atom. The edge lying highest in energy therefore corresponds to the binding energy of the K shell electron. For gamma ray energies slightly above the edge, the photon energy is just sufficient to undergo a photoelectric interaction in which a K electron is ejected from the atom. For gamma ray energies slightly below the edge, this process is no longer energetically possible and therefore the interaction probability drops abruptly. Similar absorption edges occur at lower energies for the L, M,... electron shells of the atom.

3.2.2. Compton Scattering

The interaction process of Compton scattering takes place between the incident gamma ray photon and an electron in the absorbing material. It is most often the predominant interaction mechanism for gamma ray energies typical of radioisotope sources.

In Compton scattering, the incoming gamma ray photon is deflected through an angle θ with respect to its original direction. The photon transfers a portion of its energy to the electron (assumed to be initially at rest), which is then known as a recoil electron. Because all angles of scattering are possible, the energy transferred to the electron can vary from zero to a large fraction of the gamma ray energy.

The expression which relates the energy transfer and the scattering angle for any given interaction can be simply derived by writing simultaneous equations for the conservation of energy and momentum. Using the symbols defined in the sketch below:

Fig. 3-3 Compton Scattering⁸

it can be shown that

$$h\nu' = \frac{h\nu}{1 + \frac{h\nu}{m_0c^2}(1 - \cos\theta)} \quad (3-7)$$

where m_0c^2 is the rest mass energy of the electron (0.511 MeV). For small scattering angles θ , very little energy is transferred and the secondary photon possesses almost the same energy as the incoming photon. Some of the original energy is always retained by the incident photon, even in the extreme of backscattering with large angles like $\theta = \pi$. Thus for no scattering angle the gamma ray energy can be fully absorbed with just one interaction.

The probability of Compton scattering per atom of the absorber depends on the number of electrons available as scattering targets, and therefore increases linearly with Z . The dependence on gamma ray energy is illustrated in Fig. 3-2 for the case of sodium iodide and generally falls off gradually with increasing energy.

3.2.3. Pair production

If the gamma ray energy exceeds twice the rest mass energy of an electron (1.02 MeV), the process of pair production is energetically possible. As a practical matter, the probability of this interaction remains very low until the gamma ray energy approaches twice this value, and therefore pair production is predominantly confined to high energy gamma rays. In the interaction (which must take place in the coulomb field of a nucleus), the gamma ray photon disappears and is replaced by an electron-positron pair. All the excess energy carried in by the photon above the 1.02 MeV required to create the pair goes

⁸ (Knoll, 1979), p. 65

into kinetic energy shared by the positron and electron. Because the positron will subsequently annihilate after slowing down in the absorbing medium, two annihilation photons are normally produced as secondary products of the interaction. The subsequent fate of this annihilation radiation has an important effect on the response of gamma ray detectors, as described in Chapter 10. No simple expression exists for the probability of pair production per nucleus, but its magnitude varies approximately as the square of the absorber atomic number. The importance of pair production rises sharply with energy, as indicated in Fig. 3-2.

The relative importance of the three processes described above for different absorber materials and gamma ray energies is conveniently illustrated in Fig. 3-4. The line at the left represents the energy at which photoelectric absorption and Compton scattering are equally probable as a function of the absorber atomic number. The line at the right represents the energy at which Compton scattering and pair production are equally probable. Three areas are thus defined on the plot within which photoelectric absorption, Compton scattering, and pair production each predominate.

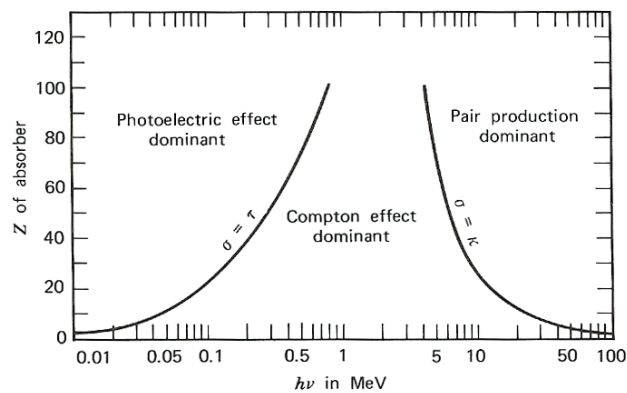


Fig. 3-4 The relative importance of the three major types of gamma ray interactions. The lines show the values of Z and $h\nu$ for which the two neighbouring effects are just equal.⁹

3.2.4. Gamma ray attenuation

Each of the three interaction processes previously explained removes the gamma ray photon from the beam either by absorption or by scattering away from the detector direction, and can be characterized by a fixed probability of occurrence per unit path length in the absorber. The sum of these probabilities is

⁹ (Knoll, 1979), p. 67

simply the probability per unit path length that the gamma ray photon is removed from the beam:

$$\mu = \tau(\text{photoelectric}) + \sigma(\text{Compton}) + \kappa(\text{pair}) \quad (3-8)$$

and is called the *linear attenuation coefficient*. The number of transmitted photons I is then given in terms of the number without an absorber I_0 as

$$\frac{I}{I_0} = e^{-\mu t} \quad (3-9)$$

The gamma ray photons can also be characterized by their mean free path λ , defined as the average distance traveled in the absorber before an interaction takes place. Its value can be obtained from

$$\lambda = \frac{\int_0^{\infty} x e^{-\mu x} dx}{\int_0^{\infty} e^{-\mu x} dx} = \frac{1}{\mu} \quad (3-10)$$

and is simply the reciprocal of the linear attenuation coefficient. Typical values of λ range from a few mm to tens of cm in solids for common gamma ray energies.

Use of the linear attenuation coefficient is limited by the fact that it varies with the density of the absorber, even though the absorber material is the same. Therefore, the *mass attenuation coefficient* is much more widely used, and is defined as

$$\text{Mass attenuation coefficient} = \frac{\mu}{\rho} \quad (3-11)$$

where ρ represents the density of the medium. For a given gamma ray energy, the mass attenuation coefficient does not change with the physical state of a given absorber. For example, it is the same for water whether present in liquid or vapour form. The mass attenuation coefficient of a compound or mixture of elements can be calculated from:

$$\left(\frac{\mu}{\rho}\right)_c = \sum_i w_i \left(\frac{\mu}{\rho}\right)_i \quad (3-12)$$

where the w_i factors represent the weight fraction of element i in the compound or mixture.

In terms of the mass attenuation coefficient, the attenuation law for gamma rays now takes the form:

$$\frac{I}{I_0} = e^{-\left(\frac{\mu}{\rho}\right)\rho t} \quad (3-13)$$

The product ρt , known as the *mass thickness* [kg/m^2] of the absorber, is now the significant parameter which determines its degree of attenuation. The thickness of absorbers used in radiation measurements is therefore often measured in mass thickness rather than physical thickness, because it is a more fundamental physical quantity.

The mass thickness is also a useful concept when discussing the energy loss of charged particles and fast electrons. Even if different absorber materials are involved, a particle will encounter about the same number of electrons passing through absorbers of equal mass thickness. Therefore, the stopping power and range, when expressed in units of ρt , are roughly the same for materials that do not differ greatly in Z .

The assumptions we have just made are being made under a so called "good geometry". The essential characteristic is that only gamma rays from the source which escape interaction in the absorber can be counted by the detector. Real measurements are often carried out under different circumstances in which the severe collimation of the gamma rays is absent and simply not possible.

Now the detector either can respond to gamma rays directly from the source, or to gamma rays that reach the detector after having scattered in the absorber. Many types of detectors will be unable to distinguish between these two possibilities, so that the measured detector signal will be larger than that recorded under equivalent "good geometry" conditions. The conditions that lead to the simple exponential attenuation of Equation (3-9) are therefore violated in this "broad beam" or "bad geometry" measurement because of the additional contribution of the scattered gamma rays. This situation is usually handled by replacing Equation (3-9) by the following:

$$\frac{I}{I_0} = B(t, E_\gamma)e^{-\mu t} \quad (3-14)$$

where the factor $B(t, E_\gamma)$ is called the *build-up factor*. The exponential term is retained to describe the major variation of the gamma ray counting rate with absorber thickness, and the build-up factor is introduced as a simple multiplicative correction. The magnitude of the build-up factor depends on the type of gamma ray detector used, because this will affect the relative weight given to the direct and scattered gamma rays.

The build-up also depends on the specific geometry of the experiment. As a rough rule of thumb, the build-up factor for thick slab absorbers tends to be about equal to the thickness of the absorber measured in units of mean free path of the incident gamma rays, provided the detector responds to a broad range of gamma ray energies.

3.3. Detectors for gamma spectroscopy

This chapter offers a brief overview of the different detector types used for radiation detection and covers the advantages and disadvantages of each type. Radiation detection instruments currently used are either gas filled detectors or solid state detectors (e.g. scintillation or semiconductor detectors).

The second part is dedicated to the performance indicators of detector hardware and their impact on the practical application in the field.

3.3.1. Gas filled detectors

A gas filled detector is usually cylindrical in shape, with an outer wall and a central electrode well insulated from each other. The wall is usually made of tissue equivalent material for ionization chamber detectors and of brass or copper for other types of detector.

Depending upon the design of the gas filled detector and the voltage applied between the two electrodes, the detector can operate in one of three regions, shown in Fig. 4.1 (i.e. the ionization region B, proportional region C or Geiger-Müller (GM) region E). Regions of recombination and of limited proportionality in the 'signal versus applied voltage' plot (regions A and D, respectively, in Fig. 4.1) are not used for survey meters.

The gas is usually a non-electronegative gas in order to avoid negative ion formation by electron attachment, which would increase the collection time in the detector, thus limiting the dose rate that can be monitored. The increase in

charge collection time results from the relatively slow mobility of ions, which is about three orders of magnitude smaller than that of electrons. Noble gases are generally used in these detectors.

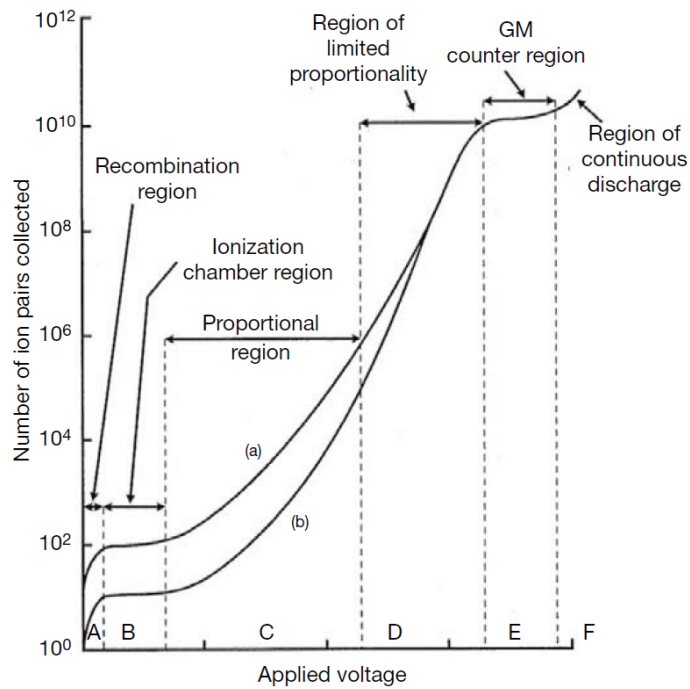


Fig. 3-5 Regions of operation for a gas filled detector¹⁰.

Region A represents the recombination region, region B the ionization region, region C the proportionality region, region D the region of limited proportionality and region E the GM region. Curve (a) is for 1 MeV particles, curve (b) for 100 keV particles.

β/γ - survey meters have a thin end window to register weakly penetrating radiation. The gamma efficiency of these detectors is only a few per cent (as determined by the wall absorption), while the beta response is near 100% for beta particles entering the detector.

Owing to their high sensitivity, the tubes of GM based gamma monitors are smaller in size than ionization chamber type detectors.

Depending upon the electronics used, detectors can operate in a “pulse” mode or in the “mean level” or current mode. Proportional and GM counters are normally operated in the pulse mode.

¹⁰ http://www-naweb.iaea.org/nahu/dmrip/pdf_files/Chapter4.pdf, April 10th, 2010

Owing to the finite resolving time (the time required by the detector to regain its normal state after registering a pulse), these detectors will saturate at high intensity radiation fields. Ionization chambers operating in the current mode are more suitable for higher dose rate measurements.

The three main types of gas-filled detectors are:

3.3.1.1. Ionization chambers

In the ionization region the number of primary ions of either sign collected is proportional to the energy deposited by the charged particle tracks in the detector volume. Owing to the linear energy transfer (LET) differences, the particle discrimination function can be used (see Fig. 3-1 Fig. 3-5). Build-up caps are required to improve detection efficiency when measuring high energy photon radiation, but they should be removed when measuring lower energy photons (10–100 keV) and β particles.

3.3.1.2. Proportional chambers

In the proportional region there is an amplification of the primary ion signal due to ionization by collision between ions and gas molecules (charge multiplication). This occurs when, between successive collisions, the primary ions gain sufficient energy in the neighbourhood of the thin central electrode to cause further ionization in the detector. The amplification is about 10³–10⁴-fold. Proportional counters are more sensitive than ionization chambers and are suitable for measurements in low intensity radiation fields. The amount of charge collected from each interaction is proportional to the amount of energy deposited in the gas of the counter by the interaction.

3.3.1.3. Geiger-Müller (GM) Counters

The discharge spreads in the GM region throughout the volume of the detector and the pulse height becomes independent of the primary ionization or the energy of the interacting particles. In a GM counter detector the gas multiplication spreads along the entire length of the anode. Gas filled detectors cannot be operated at voltages beyond the GM region because they continuously discharge.

Owing to the large charge amplification (nine to ten orders of magnitude), GM survey meters are widely used at very low radiation levels (e.g. in areas of public

occupancy around radiotherapy treatment rooms). They are particularly applicable for leak testing and detection of radioactive contamination.

GM counters exhibit strong energy dependence at low photon energies and are not suitable for use in pulsed radiation fields. They are considered indicators of radiation, whereas ionization chambers are used for more precise measurements.

GM detectors suffer from very long dead times, ranging from tens to hundreds of milliseconds. For this reason, GM counters are not used when accurate measurements are required of count rates of more than a few hundred counts per second. A portable GM survey meter may become paralysed in a very high radiation field and yield a zero reading. Ionization chambers should therefore be used in areas where radiation rates are high.

3.3.2. Scintillation detectors

Scintillation Detectors are based on the detection of ionizing particles by the scintillation of light when those particles pass certain materials referred to as “phosphors”. The emitting light is being converted to an electrical pulse by so called photomultiplier tubes (PMT) which will be explained a little later in this text.

There are various properties which make the ideal scintillation material:

1. The incoming particle should be converted to light with a high efficiency
2. The conversion should be linear over a wide range in terms of the energy of the incoming particle.
3. The material should be transparent to the wavelength of its emitted light to increase efficiency.
4. The decay time of the excited state should be short to ensure the detection of fast signal pulses
5. The materials index of refraction should be as near to those of glass as possible to ensure efficient coupling to a photo multiplier.

Since there is no material which meets all the desired properties, one has to take account of the specific experimental needs before choosing the material.

The most common materials are inorganic scintillators like Sodium Iodide (NaI) or Lanthanum Bromide Crystals (LaBr_3) and organic-based liquid or plastic scintillators. While inorganic scintillators tend to have better light output and

linearity they are rather slow in terms of their response time. On the other hand the organic scintillators are much faster but they yield less light.

3.3.2.1. Organic Scintillators

Since the fluorescence properties in organic materials arise from the transitions of the energy levels in single molecules, the scintillation properties of those materials are not bound to a specific state. To be different to inorganic crystal scintillators, which require a regular crystalline lattice for the scintillation process, organic scintillators maintain their scintillation properties in either solid, solute or vaporised state.

Types of organic scintillators:

Pure Organic Crystals

The most popular organic crystal scintillators are Anthracene (highest scintillation efficiency) and Stilbene. Major problems with those materials result from the difficulty to produce large crystals and the strong dependency of the scintillation efficiency on the orientation towards the incoming particle (20-30%).

Liquid Organic Solutions

Liquid scintillators are produced by dissolving an organic scintillator in an appropriate solvent. In addition it is routine to use a wavelength shifter to adopt the emission spectrum of the scintillator for specific purposes.

Liquid scintillators are widely used when large detection volumes or special shapes of the containment are needed. One problem is the dissolving Oxygen which reduces the scintillation efficiency.

Plastic Scintillators

These types of scintillators are produced by polymerizing a proper dissolution of an organic scintillator in a proper solvent. Just like the liquid scintillators the plastic scintillators can be brought to almost every shape and are therefore extremely useful and flexible in application. Another advantage is the relatively low price and the wide range of commercial availability.

Thin Film Scintillators

Those very thin plastic scintillators ($\sim 20\mu\text{g}/\text{cm}^2$) play an important role in their application as radiation detectors.

Loaded Organic Scintillators

Due to the low Z-value of the constituents of organic scintillators (H, C, O) the photoelectric cross-section is vanishing. In order to provide a higher photoelectric cross-section for those materials one has been trying to add higher Z elements to the base-material. The most common additives are for example lead or tin which are added up to an amount of 10% of the detector material. This results in a relatively high gamma efficiency for those materials.

3.3.2.2. Inorganic Scintillators

The crystal lattice determines certain energy states for specific scintillation-materials on which the scintillation mechanisms depend. In insulator or semiconductor materials the electrons energy can only be found in two discrete energy bands (Fig. 3-6) – the valence band and the conduction band. In between those bands – in the forbidden band – electrons cannot be found in a pure crystal. Due to this fact energy absorbed from a detected particle can only lift a lattice-electron from the valence band to the conduction band. Since this energy gap is very large, the energy of the photon resulting from de-excitation of the electron in the conduction band would be too high to be visible.

To get rid of this problem, small amounts of so called “activator” atoms (impurity elements) are added to the pure crystal structure. The result is that energy levels in the forbidden band become available for the excited electrons and thus the light resulting from de-excitation is within the visible spectrum and can therefore be used for a scintillation process.

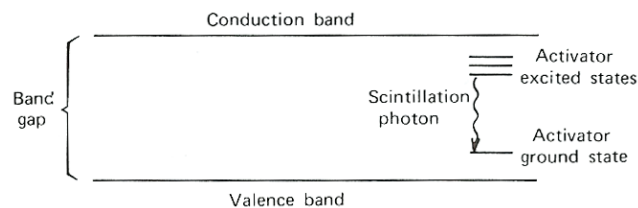


Fig. 3-6 Energy structure of an activated crystalline scintillator (Knoll, 1979)

3.3.2.3. Photo Multiplier

The whole use of scintillation detectors couldn't be realised without devices which convert the very weak light output of the scintillation material into a corresponding electrical signal: *The Photo Multiplier Tube (PM)*.

The goal is to convert the output of a few hundred photons to a corresponding electrical signal which can be further processed. TPM Tubes multiply the signal

produced by incident light by over 100 times, from which single photons can be resolved. The combination of high gain, low noise, high frequency response and a large area of collection ensure the important role of those devices in the field of radiation detection and processing. Semiconductor devices like avalanche photodiodes have replaced photomultipliers in some applications, but photomultipliers are still used in most cases.

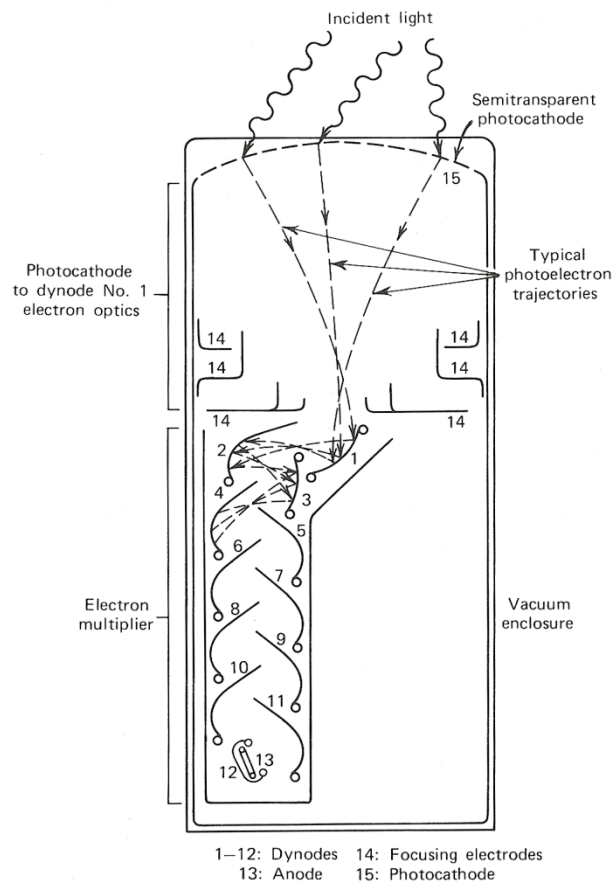


Fig. 3-7 Setup of a photo multiplier tube (Knoll, 1979)

The PM Tube is connected to a scintillation detector by a light conductor. A photocathode converts incoming photons to electrons which enter the electron multiplier, which consists of multiple dynodes. As the electrons move towards the first dynode they are accelerated by the electric field and arrive with much greater energy. When hitting the first dynode, more electrons are emitted and those are again accelerated toward the next dynode and so forth. The geometry of the dynode chain is such that an avalanche occurs with an ever-increasing

number of electrons being produced at each dynode. Finally the anode is reached where the accumulation of charge results in a sharp current pulse indicating the arrival of a photon at the photocathode. This pulse can be further processed by readout-electronics such as a multichannel-analyser (MCA).

As a semiconductor equivalent to PM tubes the *avalanche photodiodes (APD)* have been introduced. Those use the avalanche breakdown effect (known from Zener-diodes) and provide a signal multiplication similar to the PM.

The decision whether to use organic or inorganic scintillation materials has to be done under the estimation of the intended application of the detector. In the existing case of gamma spectroscopy it is more likely to use inorganic scintillators because of the higher Z-value of their constituents and the high density of the inorganic crystals.

Both devices introduced (ICx IdentiFINDER and Aspect MKC A03) use NaI(Tl) crystals as detection unit.

3.3.3. Semiconductor detectors

The huge advantage of semiconductor detectors are their very high energy resolutions compared to other solid state radiation detectors (e.g. scintillation detectors) and their reduced size compared to gas-filled detectors.

A semiconductor detector is a diode operated in reverse biased mode resulting in an enlarged depletion zone. Operated like this a semiconductor detector acts like a solid state ionisation chamber. Any particle or ionizing radiation crossing the depletion zone ionises the detector material and produces pairs of electrons and holes. Both electron and holes drift to the opposite electrode and create an output-signal which can be detected by readout electronics. The number of electron-hole pairs is proportional to the energy transmitted by the ionizing radiation to the semiconductor material.

Besides the high energy resolution capabilities, semiconductor detectors convince with many other nice characteristics like reduced ionisation energy, fast timing characteristics, compact size and an effective thickness, which can be varied in terms of the application.

On the other hand semiconductor detectors are very expensive and require sophisticated cooling during operation to prevent disturbing leakage currents (noise reduction).

Semiconductor detectors are widely used in high energy physics, e.g. in vertex detectors of some huge experiments like CMS (Compact Muon Solenoid) on

CERN and, of course, for spectroscopy reasons like gamma spectroscopy in the field of nuclear security.

The most commonly used materials are Silicon and Germanium. Silicon detectors are restricted in terms of their thickness (< some millimetres) while Germanium can have a depleted, active region of some centimetres. State of the art High Purity Germanium detectors (HPGe) can be stored at room temperature and need only a short time to be cooled down to operation temperature. Meanwhile handheld HPGe devices allow independent in-field operation for some hours using a single battery pack.

3.3.4. Multi Channel Analyser (MCA)

Now that the detection of the event (a particle crossing the detector volume) has taken place and the electrical signal has been produced, the signal has to be processed in order to gain the desired data – the spectrum.

The amount of electrical charge is proportional to the amount of gamma-ray energy absorbed in the detector volume and the job of the electronic system is to collect the charge, measure the amount and store the data.

The output signal of the detector unit has to be processed and amplified using various amplification stages (pre-amplifier, amplifier) before entering the multichannel analyser. The pre-amplifier collects the electron-hole pairs and converts the charge to a voltage pulse. This pulse is being shaped and increased in size by the amplifier.

The multichannel analyser sorts pulses retrieved from the amplifier by pulse height, which is corresponding to the energy of the detected particle. This leads to multiple channels (energy intervals) for which the multichannel analyser counts the incoming pulses. The result is a spectrum where bins of certain energies are compared to the number of counts in the related bin. The number of bins (channels) can vary between 512, 1024, 2048, 4096 or more, depending on the desired resolution (energy/channel) and energy range covered by the whole spectrum. For example a 1024 channel spectrum at 3keV / channel would cover an energy range from zero to 3MeV – those are the settings at which most measurements in this work have been carried out.

An ideal MCA would perform the conversion of pulse height to energy level (channel) perfectly linear. In that case the correlation of channel number to energy level can be determined by two parameters, the slope/gain (Fig. 3-8) and the intercept point of the calibration line with the zero baseline (Fig. 3-9). Those

parameters can be determined by measuring the FWHM of the full energy peak of well known sources, and then fitting the calibration line accordingly.

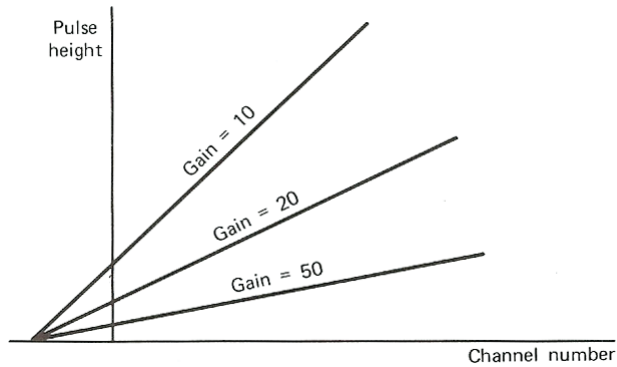


Fig. 3-8 Multichannel Analyser calibration plot for different amplifier gains (Knoll, 1979)

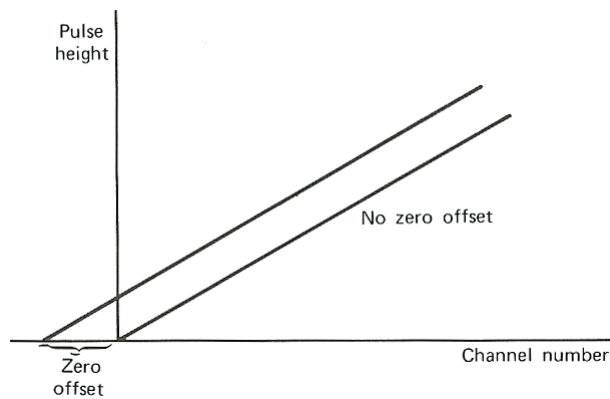


Fig. 3-9 Calibration plot for a multichannel analyser with and without zero offset (Knoll, 1979)

3.4. Gamma spectroscopy

This chapter covers specific properties of gamma spectra and the detection systems one has to take into account when doing gamma spectroscopy. Some basic concepts of the process from the spectrum to the analysis result and the nuclide identification will be described.

Another focus will be laid on the various methods for nuclide identification – the base for a proper treatment of incidents for those who work with detection devices in the field.

3.4.1. General properties of detectors and definitions

Some of the basic properties of gamma spectra and the detection system one has to deal with will be explained here.

- **Photo Peak / Full Energy Peak**

At low energies the interaction of the radiation with the detector material happens mostly by means of the photo effect. The whole energy $E_\gamma = h\nu$ of the gamma quantum is being transferred to the photo electron. The resulting line in the spectrum is called the *Photo Peak* or *Full Energy Peak* which represents the energy of the originating gamma quantum. If there are more than one radioactive materials recorded in a spectrum, there will be several full energy peaks too.

- **Compton Continuum / Compton Edge**

At higher energies the photo effect loses importance while the influence of the Compton Effect is increasing. If the gamma quanta are scattered back at an angle of $\vartheta = 180^\circ$, the whole energy (E_c) is transferred to an electron of the detector material. For any other angles $\vartheta < 180^\circ$ the scattered electrons have lower energies. As a result in the gamma spectrum, starting at E_c (the so called *Compton Edge*) down to low energies the *Compton Continuum* builds up. If the scattered gamma quantum doesn't leave the detector volume and experiences further scattering the result is additional background noise in the spectrum.

- **Back Scattering Peak**

If the Compton Scattering takes place outside of the detector material (e.g. in the radiating material itself), only the scattered gamma quanta with energies starting at $E_{BS} = E_\gamma - E_c$ can be detected. In the spectrum the so called *Back Scattering Peak* shows up at E_{BS} .

- **Single Escape Peak / Double Escape Peak**

For high energy gamma quanta the probability for the Compton Effect is reduced while the probability for pair production increases for energies higher than $E_\gamma > 2m_0c^2$. The produced electrons and positrons carry the energy difference of $\Delta E = E_\gamma - 2m_0c^2$. Since the positrons annihilate immediately with the electrons of the detector material, two gamma quanta with the energy m_0c^2 are produced. If both of them are absorbed within the detector, a peak at E_γ will show up. If one of the quanta escapes the detector, a single escape peak at $E_\gamma - m_0c^2$ occurs, if both quanta escape, a double escape peak at $E_\gamma - 2m_0c^2$ shows up.

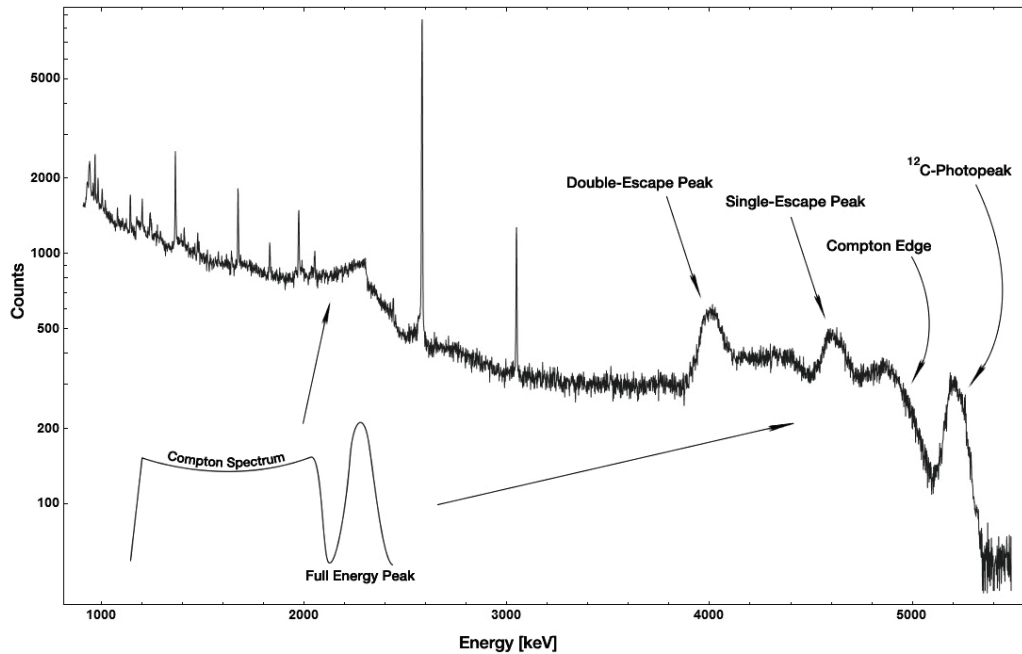


Fig. 3-10 Gamma spectrum of an Am-Be source showing Compton Spectrum, Compton Edge, Single and Double Escape peaks and the Full Energy Peak. (From <http://en.wikipedia.org/wiki/File:Am-Be-SourceSpectrum.jpg>)

- **Energy resolution / Detector resolution**

A high *energy resolution* of a detector is important to distinguish between adjacent peaks in a spectrum. Resolution in gamma spectroscopy is comparable to the resolving power in optics. Since the energies of gamma quanta are not pictured as single lines but as broadened peaks, shaped in a Gaussian distribution, the resolution is even more important to identify the single peaks.

- **FWHM**

The most common way to express the energy resolution of a detector is the "*Full Width at Half Maximum*", in short *FWHM*, which is the width of the gamma ray peak at half of the highest point on the peak distribution. It is important to give a reference to a specific gamma ray energy. For example one can have a FWHM of 10keV at 660keV and a FWHM of 30keV at 1.2MeV.

- **Detector efficiency**

The probability that a gamma quantum passing through the detector volume actually produces a count is called the *efficiency* of a detector. The higher the efficiency the lower is the time required to take a

spectrum. The surrounding of the detector (e.g. shielding) has also effects on the efficiency. Larger detectors are normally more efficient.

- **Linearity**

As mentioned before multi channel analysers should provide a most linear correlation of channel numbers to the related gamma energies. Small Non-Linearities occur at extreme pulse heights but they are rather small (<1%) for common Multichannel Analysers.

- **Dead Time**

This is the minimum time required of a detection system to record two subsequent events as separate events. The problem is that the detection system cannot count a true event because it occurs too quickly after a preceding event. This problem obviously gets more important if the count rate increases. Dead time can give errors in two different manners. First case is, that the detectors is “blocked” by an event and cannot detect the next event until the preceding one has vanished and the other possibility is, that the detector is still “live” after the detection of an event. The successive incoming event will then produce a signal as well, but the detector as well as the electronics will be unable to distinguish between those events and produce a false output (pile-up).

- **Pile-up**

Since the amplitude of the detector signal carries the main information, the “pile-up” of pulses is a serious problem. If the preceding pulse has not yet decayed to zero, the pulse height of the following pulse can be superimposed to the preceding which results in a wrong amplitude and therefore in a wrong energy-measurement. To get rid of this problem the pulses can be shaped in order to maintain the important information (maximum amplitude) and cut off possible error-sources like the long tail of a decaying pulse.

- **Background Radiation**

Every detector produces a background signal originating from natural radioactive material (e.g. rocks containing K-40, U-235,...) and cosmic radiation. Background radiation can be reduced by using proper shielding of the detection device. In any case it is important to take spectra of the background radiation in order to specify the surrounding parameters and later subtract the background from the measured

spectra to increase the minimum detectable radiation level. Sources of Background can be¹¹:

1. Natural Radioactivity of the constituent materials of the detector
 2. Natural Radioactivity of auxiliary equipment, shielding and supports near the detector unit
 3. Radiation from the earth's surface ("terrestrial radiation")
 4. Radiation in the air surrounding the detector (e.g. Radon)
 5. Primary and secondary components of cosmic radiation
- **Calibration**

Since the channel number is proportional to the detected energy, the detecting device has to be calibrated before the measurement process. Calibration is usually done using a source with well-known full energy peaks such as Cs-137 or Co-60. Since the peaks are well known, the channel-scale can easily be fitted to the energy-scale.
 - **Response Function**

A detector unit exposed to a monoenergetic radioactive source produces a differential pulse height distribution. This distribution is called the *response function* of the detector for the specific energy used.
 - **Stabilisation**

The gain from e.g. NaI(Tl) detectors and photo multiplier tubes is well known to vary with temperature. Therefore a gain stabilization algorithm has to run in the background to compensate the gain shift and guarantee a drift-free operation. The latest method to enable gain stabilization is using a thermally compensated light emitting diode and offers several advantages over designs based upon using a radioactive seed. The radioactive seed adds significant cost to the crystal and poses potential disposal problems when the crystal is no longer usable. The LED takes the signal from the temperature sensor of the detector. It then compares the LED signal from the PM tube with a temperature compensated reference signal and generates a correction voltage to automatically adjust the high voltage to provide gain stabilization.¹²

¹¹ (Knoll, 1979)

¹² <http://www.canberra.com/products/1127.asp>, 5.6.2010

4. Experimentation

This section will provide information about the various methods and algorithms to identify nuclides as well as explain the functionality of the used devices, their technical properties, benefits and failures.

The experimental part contains the setup of the measurements, the procedure and the results following a conclusion. The first experimental part describes measurements to determine the detection limits, both of the devices and the used software (IdentPro). The second interesting part deals with the ability of detection hard- and software to distinguish between harmless nuclides (e.g. medical isotopes, NORM material like chemical fertilizer) and special nuclear material (SNM) like Plutonium. This context is very important, because harmless materials may be used to mask dangerous nuclides and this has to be discovered by proper hard- and software.

4.1. Methods of locating peaks in spectra

There are some approaches to the problem of discovering appropriate peaks in a gamma spectrum and linking them to the accurate isotope. Due to difficult circumstances in the field (high radiation background, low intensities of the unknown material, unknown environmental parameters,...) those algorithms have to be very elaborate and they must provide a “grade of certainty” to the field officer in order to give him a base of decision making. (Gilmore, 2008)

4.1.1. Using Regions of Interest

Using Regions of Interest (ROIs) the error rate in the detection process can be reduced by simply telling the software “where to search for peaks”. ROIs can be defined according to channels or – even better – to energies. In some software programs like WinSPECtec even overlapping ROIs can be defined to enhance the search progress. (GBS Elektronik GmbH)

4.1.2. Derivative peak search

This commonly used method uses the first and second derivative of the fitted Gaussian function to determine the presence of peaks in the spectrum. The first derivative function changes sign as it crosses the peak centroid while the second derivative reaches a minimum at the centroids position. Furthermore one could use the third or fourth derivative function for peak search algorithms as well.

Since the spectral data of a peak is not a perfect Gaussian curve but just a histogram that approximates a Gaussian curve, the derivative function cannot be calculated. Instead one calculates the differences from the channels to their preceding ones as an approximation of the gradient.

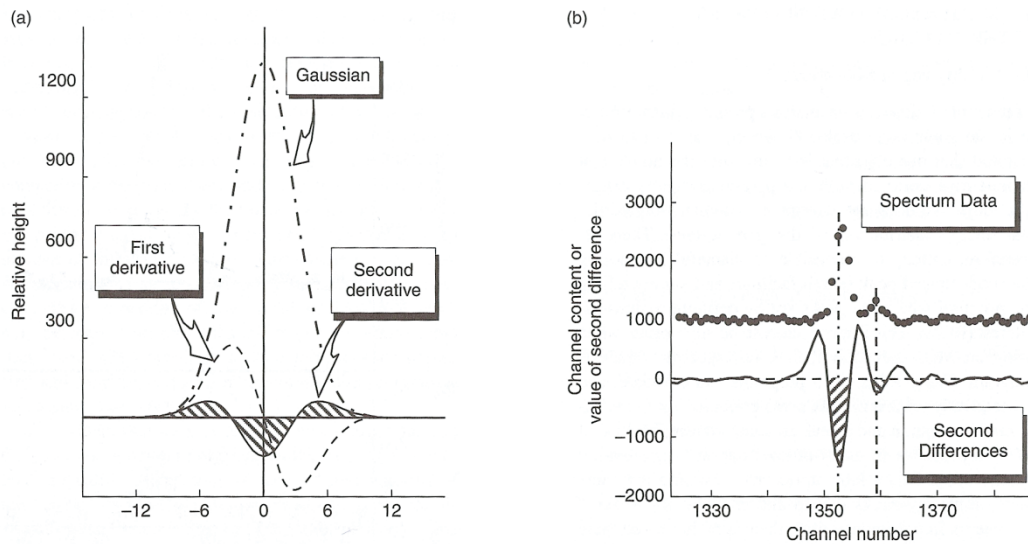


Fig. 4-1 (a) The first and second derivative function of a Gaussian peak. (b) Spectrum data and the corresponding smoothed second difference function, where negative regions indicate the presence of a peak, (Gilmore, 2008)

4.1.3. Correlation method

Using this method a search function is scanned across the whole spectrum and a “correlation spectrum” is built. Over the width of the search function each spectrum count is multiplied by the corresponding point of the search function and summed up to build one point in the correlation spectrum. After subtracting the underlying continuum by applying a bias, any points in the correlation spectrum which are greater than zero represent a found peak. Hereby statistical scatter is effectively smoothed by taking more than one channel into account for calculating one point of the correlation spectrum. In order to save processor

power, the search function can be varied from a Gaussian function (high calculating power consumption) to a simple rectangle function (lower calculating power consumption).

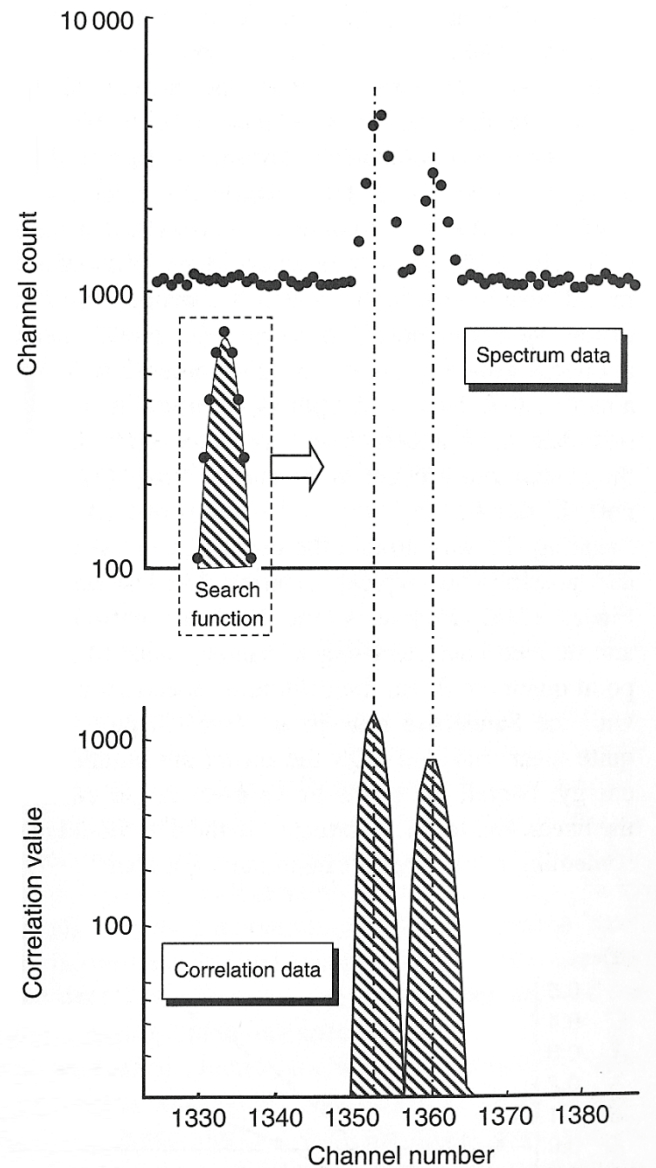


Fig. 4-2 (top) Spectrum data with a Gaussian search function (bottom) The resulting, adjusted correlation function where positive areas indicate the presence of peaks, source: Gilmore, page 187

4.1.4. Using interference coefficients

The method of interference coefficients focus on methods based on special techniques for analyzing regions of interest (ROIs). The ROI method is effective when applied to simple spectra where the principal peaks of the isotopes are quite resolved. Unfortunately, the spectra of many single and mixed-isotope sources are complex due to interference from close-lying peaks.

This method takes potential "interference" resulting from the overlap of peaks coming from different isotopes into account and uses a table of interference coefficients. This table is composed of cells that are determined, in one direction, by a set of potential isotopes in the source (candidates) and, in the other direction, by the major peaks that these isotopes will produce when present in a source. The method of analysis is to measure and equate the intensity of each major peak in a spectrum to a linear expression involving the isotopes that can contribute counts to the peak. When peaks are well resolved, the interference coefficients of non-contributing isotopes have a value of zero and the resulting equations are very simple. However, in many cases, more than one isotope may contribute to the observed intensity of a detected peak. A method of least-squares is used to solve the set of equations. The isotopes most likely responsible for the observed data are those giving positive and statistically valid results. Isotopes yielding zero, negative, or statistically unacceptable results are considered not to be present and thereafter removed from further consideration. When this occurs, a new set of equations is formed and the process repeated until only positive and statistically significant results are retained. Thereafter, the remaining isotope candidates are further screened by a number of criteria and logic tests to certify their presence. (Gunnink & Arlt, IdentPro: Isotope identification software for analyzing illicit trafficking spectra, 2001)

4.1.5. Template Matching

This method is used for example by the ICx (formerly Target) identiFINDER™ device. It correlates the measured gamma spectra with a library of referenced spectra templates stored on the device. It is obvious that the quality of the referenced spectra have a significant impact on the performance of the identification process and the identification results.

To get to proper template spectra there are two ways: The recording (measurement) of such spectra, which is tedious and expensive or the generation of the template spectra by the use of Monte Carlo simulations. (Roemer & al, 2006)

4.2. Setup and technology of used devices

The challenged devices are the Target IdentiFINDER and the Aspect MKC A03. Both devices are very much comparable, they both contain a NaI(Tl) Scintillation detector, a Geiger-Müller Tube and a ^3He neutron detector system. Both devices are currently in use by the IAEA¹³ and by public authorities of the member states. As a requirement of the IAEA both devices are equipped with two modes of operation: a simple mode, which allows access to the basic functions such as surveillance mode or identification mode and secondly an expert-mode (password-protected) which enables the full access to settings, spectra and additional features. The simple mode is intended to be used by non-expert users in terms of gamma spectroscopy, who just have to get a quick overview of a certain situation and if there is any danger arising from radioactive sources.

4.2.1. Target (ICx) IdentiFINDER™ ULTRA NGH

The Target (ICx) IdentiFINDER™ instrument is a hand-held, digital gamma spectrometer and dose rate measurement instrument, which is able to rapidly locate, accurately measure and precisely identify sources or contaminations emitting gamma radiation.

This IdentiFINDER™ ULTRA NGH includes a 1.4" (35 mm) diameter x 2" (51 mm) long NaI(Tl)¹⁴ detector plus a Geiger Müller tube, for high gamma dose rate measurements. The integrated the ^3He neutron detector enhances the IdentiFINDER's capabilities in terms of the ability to detect neutrons in the radiation field. It integrates a digital multi-channel analyzer, amplifier, high voltage power supply and memory into a lightweight, easy to use device.

The IdentiFINDER™ ULTRA incorporates a unique method of stabilization that does not require a radioactive source of any type. Stabilization of the photo multiplier tube gain is accomplished by means of a pulsed LED. Measurement of the effective NaI(Tl) crystal temperature is determined by pulse shape analysis and gain shifts due to the measured temperature changes of the crystal are automatically corrected.

In the IdentiFINDER ULTRA NGH an integrated ^{137}Cs source of about 500 Bq (15 nCi) is used for calibration and stabilization.

¹³ International Atomic Energy Agency

¹⁴ Sodium Iodide (NaI) and Thallium (TI)

Stabilisation is necessary because variations in normal operating conditions can affect the performance of radio-isotope identification devices (RIID) with scintillation detectors such as NaI(Tl). Dramatic temperature variations can affect the scintillation detector and cause instabilities and non-linearities in the electronics. High count rate changes will also impact the peak position in a spectrum. The IdentiFINDER has been developed to correct all these circumstances through different solutions for automatic calibration and continuous stabilization.



Fig. 4-3 The actual version of the ICx IdentiFINDER™ ULTRA NGH

A problem one has to deal with when working with the IdentiFINDER is, that the device only triggers the identification process, if the counts in a recorded spectrum exceed a certain threshold. That is a serious problem, in particular when working with weak sources like calibration sources. To avoid this issue, a background of K-40 has been added to all the spectra recorded by “shielding” the sources with a certain amount (18kg) of fertilizer which contains Potassium and therefore a noticeable amount of the natural occurring radioactive isotope K-40 (natural abundance: 0,012%).

4.2.1.1. Specifications

Functions	<ul style="list-style-type: none"> • dose rate display • radiation guard and alarm annunciator • source finder • gamma ray spectrometer • spectrum analyzer • nuclide identifier • dose rate logger • neutron indicator
Main detector	∅ 1.4" x 2" NaI(Tl) scintillator
Integrated ancillary detectors	Geiger-Mueller tube (GM tube) for high dose rate measurements moderated ³ He tube for neutron detection
Remote Control	RS232 interface, USB adapter setup and control including online measurements
Languages	English, French, German, Russian, Spanish
Integrated electronics	<ul style="list-style-type: none"> • Digital Multi-Channel Analyzer for main detector: <ul style="list-style-type: none"> ○ Flash ADC ○ Digital Signal Processor (DSP) ○ Enhanced Filter Coprocessor (EFCOP) • HV power supplies for main and ancillary detectors • Shaping Amplifiers and Discriminators for ancillary detectors • serial interface (RS232, USB)
Stabilization	source less ULTRA detector stabilization (LED)
Stability	±1.0% to ±1.5% (typical) -15°C to +55°C, 30 to 100,000 cps
Calibration	Automatic energy calibration and stabilization

Table 4-1 Specifications of the Target (ICx) IdentiFINDER NGH according to ICx radiation

4.2.1.2. Spectrometry System - Dose / Dose Rate Measurement

Detector type	NaI(Tl)
Shaping	Digital filter
Resolution	7.0% to 7.5% at 662 keV (NaI)

Throughput rate	> 100,000 cps
Energy range	15 keV to 3 MeV
INL, top 99%	< 0.05%
DNL, top 99%	< 0.1%
Spectrum characteristics	1024 channels linearized, calibrated, and stabilized energy scale
Spectrum memory	100 spectra at 1024 channels
Real time presets	1 s – 1,000,000 s or none
Live time presets	1 s – 1,000,000 s or none
Dose rate range (total)	0.01 μ Sv/h to 1 Sv/h; or 1 μ rem/h to 100 rem/h
Dose rate range NaI	0.01 μ Sv/h to 500 μ Sv/h
Dose rate range GM Tube	500 μ Sv/h to 1,000 mSv/h
Dose range	0.1 μ Sv to 1 Sv; or 10 μ rem to 100 rem
Alarm levels	5 adjustable preset levels

Table 4-2 Spectrometry System of the Target (ICx) IdentIFINDER NGH according to ICx radiation

4.2.1.3. Dimensions

Weight	1250 g (44 oz) including batteries and NaI(Tl) detector
Temperature range	-15 °C to +55 °C
Dimensions	235 mm x 93 mm x 75 mm (9,2" x 3,7" x 3")
Operating time	10 hours with 2500 mAh NiMH in dose rate mode
Accessories	AC adapter for stationary use and recharging powerPACK plus; RS232, USB cables and connector box; holster, carrying case, optional: earphones
Keyboard	4 soft buttons
Alarm indicators	Light (red and blue LEDs), sound and vibration
External power indicator	LED (green) on the rear side
Charger indicator	LED (orange) on the rear side

Display type	Monochrome LCD with backlight, 61 mm x 43 mm (2,4" x 1,7")
Interface	RS232 and USB
Battery pack	powerPACK plus (rechargeable)
Standard batteries	powerPACK for 4 pieces AA-size 1.2 V - 1.8 V batteries (rechargeable NiMH, NiCd or non-rechargeable Alkaline, MnO, Li)

Table 4-3 Dimension of the Target (ICx) IdentIFINDER NGH according to ICx radiation

4.2.2. Aspect MKC-A03

The hand-held Radiation Monitor MKC-A03 is being manufactured by the Russian company “Aspect - Scientific Production Center” in Dubna near Moscow.

The device offers about the same functionality as the IdentifINDER and is also in use by IAEA Safeguards and Staff of IAEA member states. It can be used for the control of nuclear and radioactive material trafficking and monitoring of wide areas. Fields of application are mobile radiological laboratories, customs checkpoints, nuclear facilities, military sites, nuclear power plants and so forth.

Stabilisation is accomplished by using a small Cs-137 source.

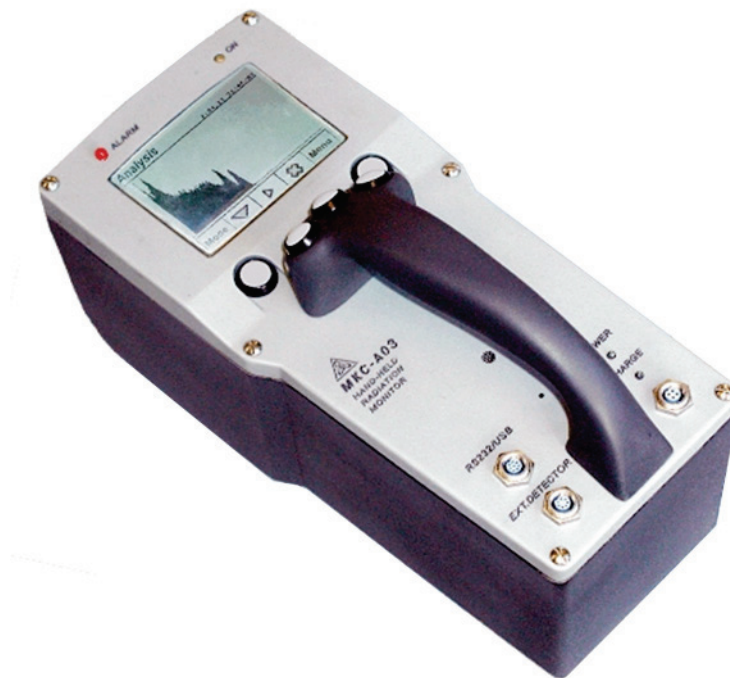


Fig. 4-4 The Aspect MKC-A03

Purpose

- Search for radioactive sources (α , β , γ , η)
- Measurement of dose rates (γ , η)
- Measurement of flux density (α , β)
- Identification of radio nuclides (γ)

Built-in detectors

- Gamma detectors: NaI(Tl), GM counter
- Neutron detector: ³He counter
- BDS-AB2 External alpha-beta detector
- BDS-06-01 External wide-wave neutron detector (upon request)

Features

- Comprehensive software for radionuclide identification
- Wide range of gamma radiation EDR measurement
- Operation of all functions with five buttons
- High resistance to mechanical damage
- Convenient connection with PC
- Wireless data transfer
- Automatic energy calibration during battery charging
- Neutron EDR measurement in a wide energy range with the external detector

Dosimetry and Radiometry

- Measurement of the equivalent dose rate
- Measurement of alpha and beta radiation flux density

Built-in Spectrometer

- Measurement of gamma spectra
- Identification of gamma-radiation sources
- Transfer of spectra to the computer
- IAEA recommended built-in library of nuclides with identification of isotopes by types: special, medical, industrial, natural
- Storage of up to 1000 measured spectra, data communication with PC through RS-232, Bluetooth (upon request)

Detected gamma radiation energies, MeV	0,05 - 3
Dimensions of NaI(Tl) scintillator crystal, mm	Ø40 x 40
Relative energy resolution, %	7,5

Maximum input load, s ⁻¹	5 x 10 ⁴
Temporary instability, %	1
Temperature instability, %/ °C, typical value	0,04

Table 4-4 Characteristics of the Aspect MKC A03

Certificates

The MKC-A03 is enrolled in the State Committee of Standards under №19945 of 04.03.2005 and in the State Register of Measuring Devices under №17406-05

4.3. Software for Identification and Simulation

4.3.1. WinSPEC

WinSPEC is software by GBS Elektronik GmbH for controlling the Multi Channel Analyser “MCA-166”. The MCA-166 and the associated software have been developed by GBS Elektronik at the research centre in Rossendorf (Dresden, Germany) for the IAEA and EURATOM¹⁵ inspectors.

There are three variants of WinSPEC according to the field of application:

- WinSPEC for Inspectors
- WinSPEC for Automation and
- WinSPEC for Technicians.

All variants are compatible to each other one, except minimal differences. The inspector’s variant comprises the functions necessary for inspections. The automation variant allows unattended measurements. That means, the program restarts the measurement after a main power failure. The technician’s variant has got three memory groups to store three spectra. It comprises all functions of the inspector’s variant and besides additional functions for manipulating and comparing of spectra. An important feature of the technician’s variant is to load a ROI set and “apply” it to another spectrum recorded with a device or simulated by other software like IdentPro.

¹⁵ The European Atomic Energy Community (EAEC or EURATOM) is an international organisation which is legally distinct from the European Union (EU), but has the same membership, and is governed by the EU's institutions.

4.3.2. IdentPro

IdentPro is a software for isotope identification by Ray Gunnink which uses the ROI method in combination with interference coefficients. The software is able to read various spectra formats. It is possible to enter various data manually to override the data provided in the spectrum file (e.g. Gain, Zero-Value, Number of channels, Energy Calibration, Absorber Data, Detector characteristics, Background spectra,...)

Another really important feature for this work was the simulation mode of IdentPro. It enables the true simulation of a user-defined real-time for a specific source-spectrum (as long as the real-time is lower or equal to the source). The simulated spectra can be saved to disk.

An outstanding feature of IdentPro is the summing of two spectra and simulating the resulting spectra at user-defined real-times. This enables the user to simulate e.g. spectra of U-235 and Ga-67 by simply summing a spectrum of U-235 with one of Ga-67. The simulation can be repeated for many times, which allows the user a conclusion of the detection capabilities of the software concerning specific ratios of the mixture U-235 and Ga-67. This feature has been used for the measurements in chapter 4.5 of this work. (Gunnink, Performance Illustrations of IdentPro)

4.3.3. Identify

Identify is a program by Jörg Brutscher also from GBS Elektronik GmbH in Germany, originally designed for the analysis of spectra measured with the MCA166 like WinSPEC. It can process spectra files in the *.spe or *.chn format and includes an efficient peak search algorithm as well as energy, efficiency and FWHM calibration.

Energy dependent efficiencies are calculated from the detector supplier data and information about source matrix and absorbers. A functionality to create custom nuclide libraries from a master nuclide library is also provided. Nuclides from a selected library can be easily assigned to the evaluated spectra. For visual proofing, calculated model spectra can be compared with the measured spectra.

The expected FWHM can be adjusted by setting the detector function. The detector and measurement geometry data can be chosen to calculate the efficiency. The "visual efficiency" functionality shows graphically the calculated energy-dependent efficiency and allows manual adjustment to measured values. Auto FWHM search tries through an iterative function to adjust the expected FWHM to the evaluated one.

The final analysis of the spectrum is executed using the peak search mechanism, which searches the spectrum for peaks, marks them and calculates centroid, peak area (with error) and FWHM. (Brutscher, Arlt, & Czock, 2001)

4.4. Discovering detection limits

The objective of this task was to establish a procedure to discover the detection limits of software and hardware used in the various fields of gamma spectroscopy and radiation detection.

It is very important to know how all components which are involved in the detection process respond to preset conditions. Do they recognise the full bandwidth of isotopes correctly? How reliable are these results anyway? Is it possible for an officer of customs, police or the fire brigade to be certain of what the devices tell him in case of an emergency? Can he be sure that the device raises an alarm at all? The reliability, the certainty of the results of an important tool like a nuclide identifying device is crucial.

In the first place it is interesting how the handheld devices compare in direct competition and secondly - but also very important – is it possible to increase the effectiveness of the hardware by implementing better processing software into the devices?

As an example for a stand-alone PC-software which copes very well with the moderate computing power of detecting devices, IdentPro is used and compared to the devices.

4.4.1. Procedure & Methodology

To meet the desired goal, the following procedure has been tested and found practicable:

1. A long term spectra of a well-known isotope is being recorded with both devices. The real time was set to 600 seconds as this would be the average time an operator would spend to identify an unknown radiating object. The identification process of the devices was not triggered but the spectrum was saved for further processing.
2. The originally recorded spectrum was “scaled” to a shorter real-time (for example 100 seconds) in a next step using IdentPro. IdentPro is able to statistically simulate a shorter runtime of a given spectrum and then trying to identify the involved isotopes. This process can be carried out in batch mode. The recognition results for each cycle are written to a

text-file which can be processed further. To get an idea of the detection abilities of IdentPro, 500 cycles have been batched and stored.

3. If IdentPro discovers the Isotope in at least 99% of the simulations, the resolved real-time (e.g. 100s) is being verified with the detecting device. In case of the IdentiFINDER this has been done in an automated mode using a batch file (Appendix B: IdentiFINDER Batch File). The MKC A03 is not able to batch the identification process so this had to be done manually.
4. In case that IdentPro could not meet the 99% requirement or the resolved real-time cannot be verified by the device, the simulated real-time has been increased and the process has been started over again.

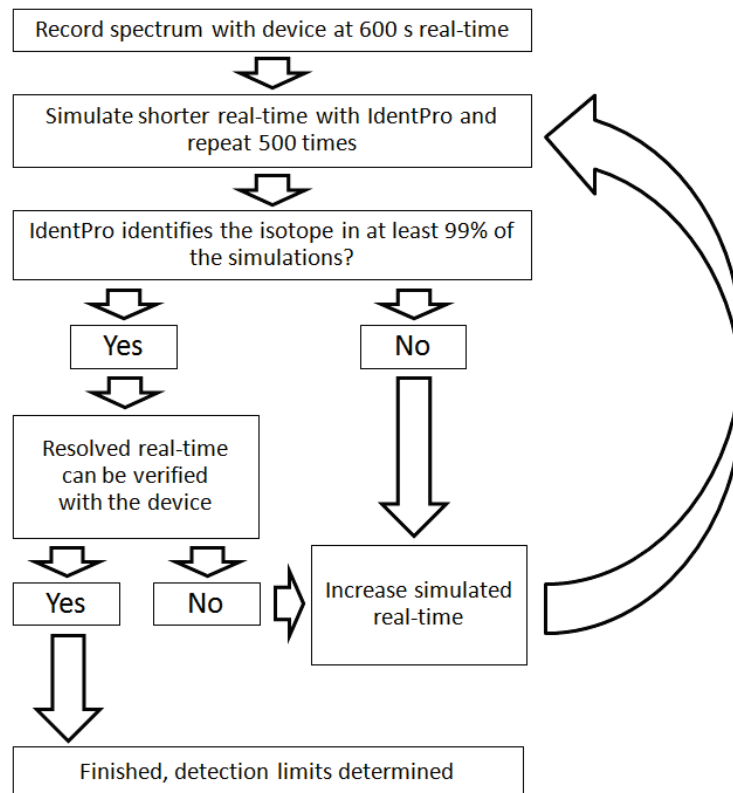


Fig. 4-5 Flow chart of the procedure to resolve the detection limits of the devices and IdentPro

The results should give an overview of how reliable the detection results from the examined devices are and if there are big differences in the detection properties depending on device and radioactive source. If some sources cannot even be identified when using the full 600 seconds real-time spectra, this points

out a serious failing of the device and should be documented and reported to the manufacturer.

4.4.2. Setup

The measurements have been carried out in the Nuclear Security Lab at the Vienna International Centre (“UNO City”).

Sources in use were calibration sources and a piece of depleted uranium. Depleted Uranium is a waste product of the uranium enrichment process and contains a lower percentage of fissile uranium (U^{234} and U^{235}) than the natural uranium ore.

Source	Serial Number	Activity Oct. 2005 [Bq]
Cs-137	3	316206
Am-241	2	321380
Eu-152	4	234300
Ba-133	2	176226
Co-60	2	107302
U-235 90% enriched	N-17	753000
Am-241	02-06	406000
Ra-226	-	unknown
depleted Uranium	-	unknown

Table 4-5 Sources at the IAEA nuclear security lab, Vienna International Centre (VIC)

Since the sources were rather weak, a box of fertilizer had to be used in order to ensure proper spectrum recording of the Target IdentIFINDER (Fig. 4-6).

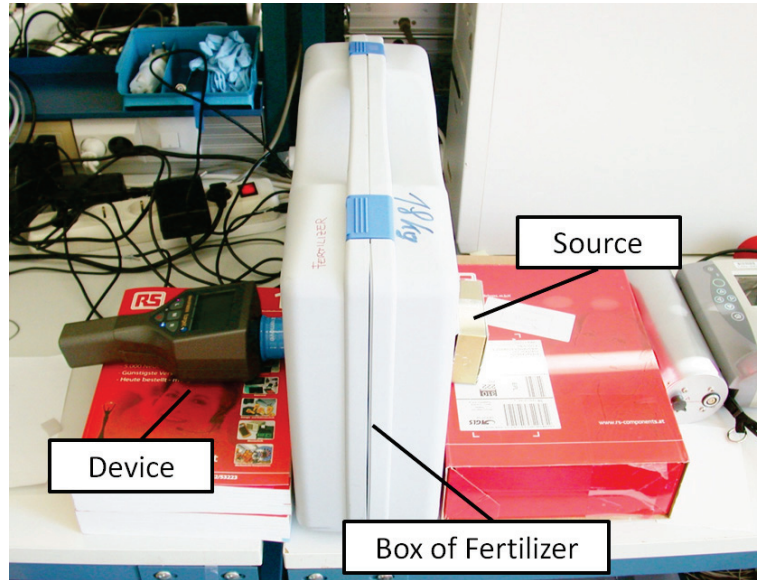


Fig. 4-6 Setup for recording 600 second real-time spectra with Target IdentiFINDER and MKC A03

The devices in use have been the IdentiFINDER ULTRA (Serial# 3367-2) and the MKC A03 (Serial# 0016/04).

Calibration of the devices has been done using a Cs-137 source

IdentPro has been used in expert mode which enables the simulation mode and allows the manipulation of some factors like the sensitivity parameters. Settings for this experiment:

- Sensitivity parameters
 - Detection sensitivity: 1
 - Change gain by: 0%
- Gain (amplification factor): 3
- Zero (defines the 0-value): 0
- “Use interface values for gain/zero”

IdentPro tries to identify all isotopes in a certain spectrum. The value of certainty according the identification is expressed on a scale from zero to 10 and is reflected in the evaluation screen of IdentPro as little “-” next to the identified isotopes. For example “ ^{133}Ba - -” indicates a detection of Ba-133 with a certainty of 20%.

The following figures illustrate the process of simulating a shorter real-time with IdentPro. Fig. 4-7 shows an original spectrum, recorded with the target IdentiFINDER. The second spectrum (Fig. 4-9) has been simulated using the

simulation mode of IdentPro (Fig. 4-8). This process is being repeated for 500 times and the results of each analysis are stored in a text file which can be easily processed afterwards with MS Excel.

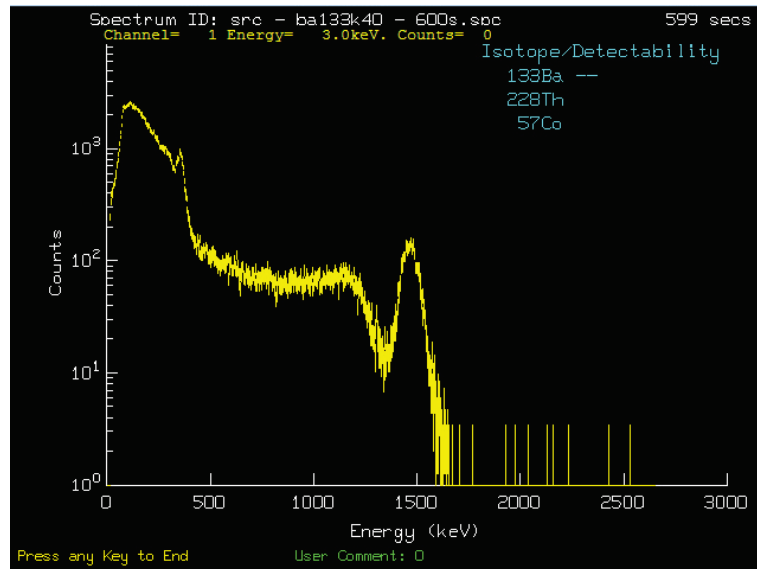


Fig. 4-7 Screenshot of Ba-133 spectrum with K-40 background recorded with the Target IdentIFINDER displayed in IdentPro: real-time of this spectrum: 600 seconds

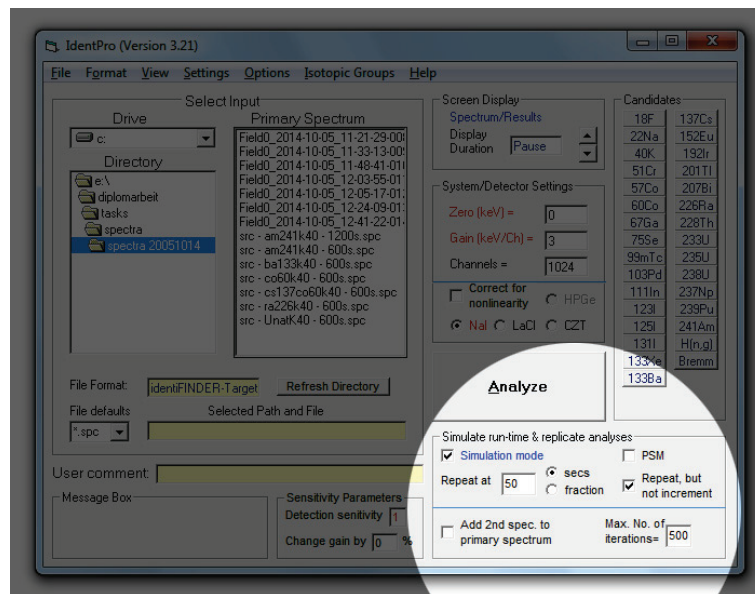


Fig. 4-8 Simulation Mode of IdentPro to simulate shorter real-times. In this case a spectrum with 50 seconds real-time is being simulated for 500 times

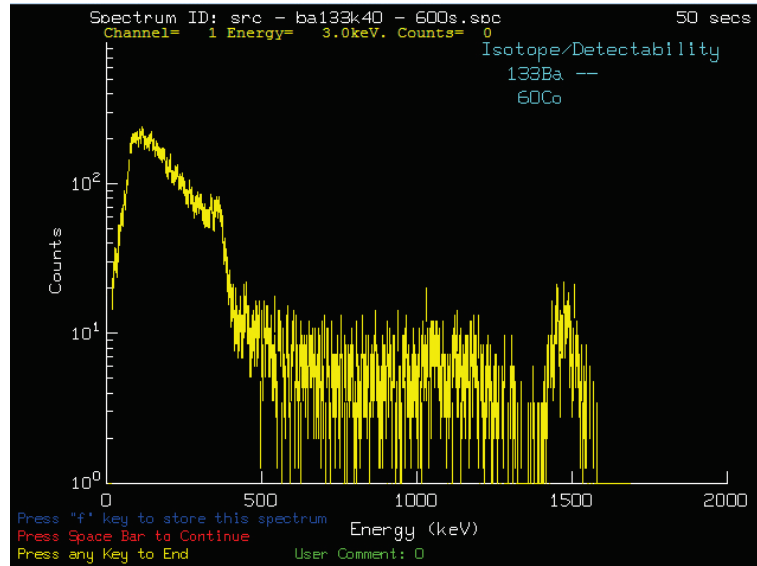


Fig. 4-9 Simulated spectrum with a real-time of 50 seconds based on an original Ba-133 spectrum (Fig. 4-7)

In this case, IdentPro can identify the radiation source as Ba-133 with a certainty of 20%. If the radiation source is recognised in more than 99% out of the 500 samples, the determined real-time – in this case 50 seconds – is going to be verified again by the device.

It is possible to automate the detection process of the Target IdentifINDER. Therefore the device is connected to a PC and controlled by the PC-Software “WinTMCA” from Target by using a batch file. A sample batch file can be found in “Appendix B: IdentifINDER Batch File”.

The results screen may look like Fig. 4-10. In this case the tested real-time is 120 seconds. The values following the “ana” command represent the identified isotopes. For example “7 NORM K-40” means that, with a certainty of 70% the isotope K-40 from the nuclide library (**Fehler! Verweisquelle konnte nicht gefunden werden. Fehler! Verweisquelle konnte nicht gefunden werden.**, page

```

OK: lpr MyPreset
OK: adcon
OK: ana 7 NORM K-40
OK: spc
OK: lpr MyPreset
OK: adcon
OK: ana 6 NORM K-40 - Ind. Ba-133
OK: spc
OK: lpr MyPreset
OK: adcon
OK: ana 7 NORM K-40 - Ind. Ba-133
OK: spc
OK: lpr MyPreset
OK: adcon
OK: ana 6 NORM K-40 4 Ind. Ba-133
OK: spc
OK: lpr MyPreset
OK: adcon
OK: ana 7 NORM K-40 - Ind. Ba-133
OK: spc
OK: lpr MyPreset
OK: adcon
OK: ana 8 NORM K-40 - Ind. Ba-133
OK: spc
OK: lpr MyPreset
OK: adcon
OK: ana 7 NORM K-40
OK: spc
OK: lpr MyPreset
OK: adcon
OK: ana 8 NORM K-40 4 Ind. Ba-133
OK: spc
OK: lpr MyPreset
OK: adcon
OK: ana 6 NORM K-40 - Ind. Ba-133
OK: spc
OK: lpr MyPreset
OK: adcon

```

Fig. 4-10 Screenshot WinTMCA: The results from the batched analyses process of the IdentifINDER. In this case the tested real-time is 120 seconds.

Fehler! Textmarke nicht definiert.) “Norm” has been found. It is obvious that the desired Ba-133 has not been detected in some cases so that’s why the real-time had to be increased and the simulation and verification process started over again.

4.4.3. Results

The measurements have been carried out for both devices. All spectral data as well as the results output text-file of IdentPro have been stored.

Explanation of terms:

- “repeated @ [s]” indicates the real-time simulated by IdentPro
- “not detected [%]” percentage of simulation which were not properly detected
- “coi [%]” certainty of identification
- “repetitions” number of verification measurements with the device
- “dose rate” dose-rate on device when recording the original spectra

4.4.3.1. Target (ICx) IdentiFINDER

Cs-137

Caesium 137 has been detected by the device and IdentPro without any problems. The simulated real-time has been decreased to as low as 50 seconds.

Source-spectrum: “src - cs137k40 - 300s.spc”

Real-time: 300 seconds

Dose-rate: 0,72 μ Sv/h

Simulation with IdentPro			Verification with IdentiFINDER		
repeated @ [s]	not detected [%]	coi [%]	repetitions	not detected [%]	coi [%]
50	0,00%	100,00%	20	0,00%	79,00%

Table 4-6 Results for Cs-137, IdentPro / Target IdentiFINDER

Depleted Uranium

Depleted Uranium was properly identified by IdentPro with no losses at a simulated real-time of 100 seconds. At this specified real-time it was not possible

to verify the results with the IdentiFINDER. The real-time had to be increased almost up to the original real-time but even then 10% of the measurements didn't lead to a proper identification.

Source-spectrum: "src - udepk40 - 600s.spc"

Real-time: 600 seconds

Dose-rate: 0,71 $\mu\text{Sv/h}$

Simulation with IdentPro			Verification with IdentiFINDER		
repeated @ [s]	not detected [%]	coi [%]	repetitions	not detected [%]	coi [%]
70	4,20%	13,25%			
90	0,20%	14,77%			
100	0,00%	13,16%	10	50,00%	< 10%
240	0,00%	~30,00%	10	40,00%	< 10%
500	0,00%	~40,00%	20	10,00%	~10,00%

Table 4-7 Results for depleted Uranium, IdentPro / Target IdentiFINDER

Ba-133

While IdentPro delivered a failure-free identification result of Barium 133 at a real-time of 120 seconds, the IdentiFINDER couldn't validate this time. Only at a threefold real-time of 360 seconds the device managed to detect Ba-133 in 100% of the measurements. What was interesting to observe is the decreasing certainty of identification in IdentPro while the simulated real-time is increasing.

Source-spectrum: "src - ba133k40 - 600s.spc"

Real-time: 600 seconds

Dose-rate: 0,32 $\mu\text{Sv/h}$

Simulation with IdentPro			Verification with IdentiFINDER		
repeated @ [s]	not detected [%]	coi [%]	repetitions	not detected [%]	coi [%]
100	1,20%	21,60%			
120	0,00%	22,25%	10	20,00%	<10%
240	0,00%	17,30%	20	15,00%	17,64%
360	0,00%	17,44%	20	0,00%	16,00%
480	0,00%	12,10%	20	0,00%	~20%

Table 4-8 Results for Ba-133, IdentPro / Target IdentIFINDER

Eu-152

IdentPro managed to detect Europium 152 without errors at a simulated real-time of 170 seconds. This time could be verified by the IdentIFINDER without any difficulty. The certainty of identification for IdentPro couldn't be increased by raising the simulated real-time and with a value of 10% it was much lower than the corresponding value of the IdentIFINDER (~56%).

Source-spectrum: "src - eu152k40 - 600s.spc"

Real-time: 600 seconds

Dose-rate: 0,61 μ Sv/h

Simulation with IdentPro			Verification with IdentIFINDER		
repeated @ [s]	not detected [%]	coi [%]	repetitions	not detected [%]	coi [%]
150	1,60%	10,00%			
170	0,00%	10,00%	20	0,00%	56,50%
200	0,00%	10,00%			

Table 4-9 Results for Eu-152, IdentPro / Target IdentIFINDER

Ra-226

It took 300 seconds real-time simulations to ensure a proper detection of IdentPro. As with Eu-152 the certainty of detection was lower for the software product than for the IdentIFINDER and could not be enhanced through increasing the real-time.

Source-spectrum: "src - ra226k40 - 600s.spc"

Real-time: 600 seconds

Dose-rate: 0,40 μ Sv/h

Simulation with IdentPro			Verification with IdentIFINDER		
repeated @ [s]	not detected [%]	coi [%]	repetitions	not detected [%]	coi [%]
200	0,20%	10,00%			
300	0,00%	10,28%	20	0,00%	28,00%
360	0,00%	10,72%			

420	0,00%	11,60%		
-----	-------	--------	--	--

Table 4-10 Results for Ra-226, IdentPro / Target IdentIFINDER

Several other mixed spectra of Potassium (K-40) and various isotopes have been tested, but since IdentPro was not able to identify the available isotopes at the full real-time of 600 and sometimes even 1200 seconds with a certainty of 100% in 500 simulations. The simulation of shorter real-times as well as the verification part didn't take place in those cases.

isotope mixture	source spectrum	real-time	dose-rate
Am-241 K-40	src - am241k40 - 600s.spc	600s	0,28μSv/h
Am-241 K40	src - am241k40 - 1200s.spc	1200s	0,28μSv/h
Co-60 K-40	src - co60k40 - 600s.spc	600s	0,65μSv/h
U-235 K-40	src - u235k40 - 3000s.spc	3000s	0,27μSv/h
Co-60 Cs-137 K-40	src - cs137co60k40 - 600s.spc	600s	0,70μSv/h
U nat K-40	src - unatk40 - 600s.spc	600s	0,29μSv/h

Table 4-11 Mixtures of isotopes which couldn't be identified by IdentPro

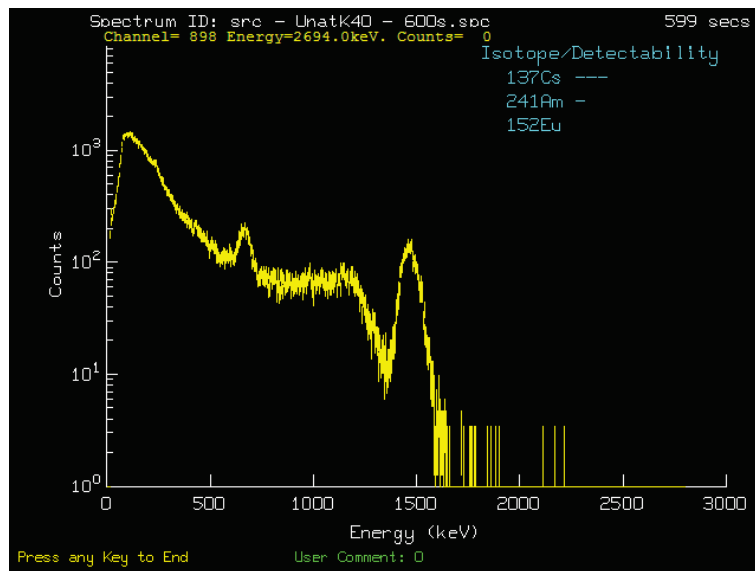


Fig. 4-11 Spectrum of natural uranium which was by mistake identified as Cs-137 | Am-241 | Eu-152

4.4.3.2. MKC A03

When taking the measurements for the 600 seconds spectra, problems with the calibration of the device occurred. Calibrating using the docking station (Cs-137 source) of the MKC A03 did not help. The Cs-137 photo peak was located at 621keV which was not acceptable for further measurements and simulations with IdentPro.

Manual calibration directly on the device (“menu” > “setup” > “analysis” > “energy calibration” > “turning a&b” > “turning a”) finally performed well and the Cs-137 peak settled at 662 keV.

All addressed spectra have been transferred to the computer using the “full range linearization” option of the MKC A03.

Some other issues which occurred during measurement:

1. The MKC Manager Software stores a real-time of 136 seconds in the spectrum data, even though the measuring time was 600 seconds. Due to this the real-time of some spectra had to be manually corrected in the spectrum file.
2. It is not possible to set the real-time on the MKC device to a time lower than 25 seconds. This would be useful / a must for evaluation purposes.
3. The MKC A03 – in contrast to the Target (ICx) IdentIFINDER does not provide an indicator for the certainty of identification on the device.

Cs-137

Even though IdentPro identified Cs-137 without any problems at a simulated real-time of 5 seconds, it was not possible to verify this value with the MKC due to the mentioned issue of a minimum real-time of 25 seconds. At this real-time it was no problem for the MKC A03 to verify the results of IdentPro.

Source-spectrum: “s0035.spc”

Real-time: 600 seconds

Dose-rate: 0,43 μ Sv/h

Simulation with IdentPro			Verification with MKC A03		
repeated @ [s]	not detected [%]	coi [%]	repetitions	not detected [%]	coi [%]
5	0,00%				
10	0,00%	91,00%			
25	0,00%	100,00%	20	0,00%	-

Table 4-12 Results for Cs-137, IdentPro / Aspect MKC A03

Depleted Uranium

The down-scaled spectrum of depleted uranium was perfectly identified by IdentPro at a real-time of 30 seconds (but with a low certainty of identification). The MKC device was not to proof this real-time. Only after increasing the real-time for about 16 times, a certain identification of depleted uranium with the MKC A03 could be achieved.

Source-spectrum: "s0036.spc"

Real-time: 600 seconds

Dose-rate: 0,39 μ Sv/h

Simulation with IdentPro			Verification with MKC A03		
repeated @ [s]	not detected [%]	coi [%]	repetitions	not detected [%]	coi [%]
30	0,00%	11,30%	5	100,00%	-
360	0,00%	20,00%	6	50,00%	-
420	0,00%	29,40%	10	20,00%	-
480	0,00%	30,00%	20	0,00%	-

Table 4-13 Results for depleted uranium, IdentPro / Aspect MKC A03

Ba-133

The real-time of this spectrum had to be corrected as mentioned above. It was interesting to observe, that IdentPro was not at all able to identify Ba-133 from the original spectrum even though the significant peaks fitted perfectly (figure XY). The results are therefore non-existent, but nevertheless it was tested if the device could identify the nuclide properly. The MKC A03 was not able to find the correct isotope in 15% of the measurements.

Source-spectrum: "s0054_corrected_rt.spc"

Real-time: 600 seconds

Dose-rate: 0,23 μ Sv/h

Simulation with IdentPro			Verification with MKC A03		
repeated @ [s]	not detected [%]	coi [%]	repetitions	not detected [%]	coi [%]
300	-	-			
480	-	-	20	15,00%	-
540	-	-	20	15,00%	-

Table 4-14 Results for Ba-133, IdentPro / Aspect MKC A03

Eu-152

Even at only 20 seconds real-time, IdentPro managed to identify clearly the Eu-152 isotope in 100% of the simulations with a certainty of identification of about 30%. However, it was not possible to verify those values with the MKC A03, because the device was not able to identify Eu-152. Even though there were all relevant peaks of the isotope present, clearly visible with the naked eye and the real-time was set to 600 seconds, no identification results were presented. The recorded spectrum with the full real-time is shown in Fig. 4-12 to illustrate this problem. The runtime of the source spectrum had to be corrected in this case.

Source-spectrum: "s0038_corrected_rt.spc"

Real-time: 600 seconds

Dose-rate: 0,47 μ Sv/h

Simulation with IdentPro			Verification with MKC A03		
repeated @ [s]	not detected [%]	coi [%]	repetitions	not detected [%]	coi [%]
10	1,00%	23,70%			
20	0,00%	29,70%	20	100,00%	-
600	0,00%	90,00%	20	100,00%	-

Table 4-15 Results for Eu-152, IdentPro / Aspect MKC A03

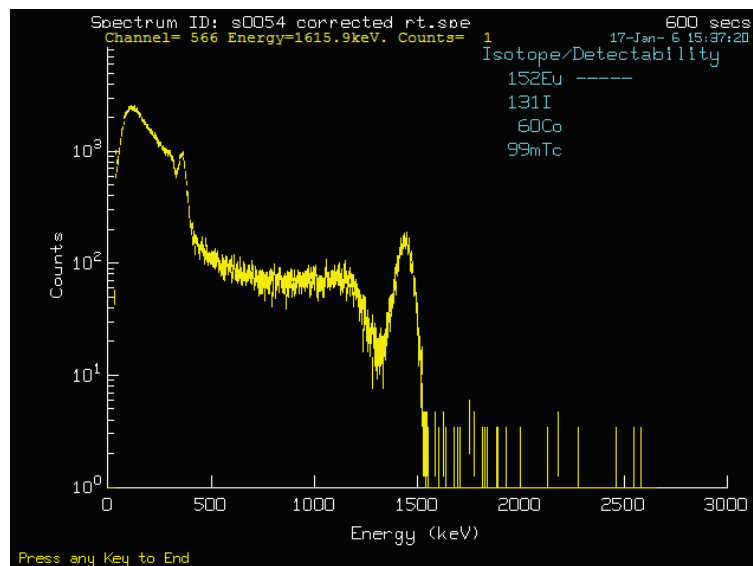


Fig. 4-12 Spectrum of Eu-152, recorded with the Aspect MKC A03. The significant peak at 344keV is easy to see, the 964keV peak a little harder.

Ra-226

IdentPro had no problem identifying the Ra-226 isotope, but it was not possible to verify those results with the MKC A03 device. Even though there were good peaks in the spectrum. The runtime in the .spc file had to be corrected again.

Source-spectrum: "s0039_corrected_rt.spc"

Real-time: 600 seconds

Dose-rate: 0,32 μ Sv/h

Simulation with IdentPro			Verification with MKC A03		
repeated @ [s]	not detected [%]	coi [%]	repetitions	not detected [%]	coi [%]
60	0,20%	12,90%			
120	0,00%	23,20%	-	-	-
480	0,00%		-	-	-

Table 4-16 Results for Ra-226, IdentPro / Aspect MKC A03

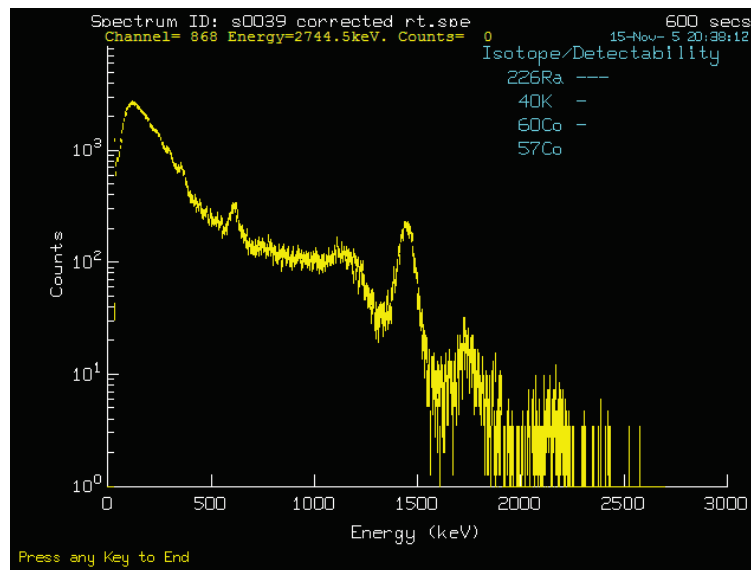


Fig. 4-13 Spectrum of Ra-226, recorded with the Aspect MKC A03. The Ra-peak at 609keV is clearly visible, but the device did not manage to identify the isotope.

As with the IdentIFINDER several other mixed spectra have been tested, but could not be identified by IdentPro at the full real-time of 600. In those cases the simulation as well as the verification part didn't take place.

4.4.4. Conclusion

As one see clearly from the results, a 600 second real-time is more than a challenge for the detecting devices in most cases of weak (or in vivo eventually shielded) sources.

Even under laboratory conditions it was sometimes not possible to get satisfactory results from the devices. Applied to the work in the field, this would lead to no or very deficient results which would be impossible to interpret for non-professional users in terms of gamma spectroscopy. Under common conditions it will not be possible to point at a suspect vehicle or person for 10 minutes or more and even if that works, one cannot be 100% sure that every threatening isotope can be clearly identified.

It is very disturbing, that the MKC A03 sometimes is not even able to resolve isotopes from a good spectrum recorded with the device itself (as with Ba-133, Eu-152 and Ra-226), while everyone can make out the relevant peaks almost with the naked eye.

In the direct competition between the two devices, the Target (ICx) IdentiFINDER shows up clearly as the more reliable device. The IdentiFINDER was able to identify almost all isotopes in less than the required 600 seconds. Only the depleted uranium caused the device some difficulties.

The role of the IdentPro software is ambivalent. On the one hand it is astonishing fast and recognises isotopes in a couple of seconds and on the other hand it cannot handle long-term spectra of americium or natural uranium. The software has enormous potential and one chance of increasing the performance of this tool is possibly the adjustment and enhancement of the correlation table of peaks.

In the end the question remains, if such handheld devices with an intrinsic limit to the size of the detector crystal are the right choice for e.g. non-professional staff at borders or customs. The devices at this very stage of development convey some sort of feeling of safety, which is not really justified.

4.5. Masking of threatening isotopes with MED isotopes

Illicit trafficking of nuclear material is a serious problem the public authorities have to cope with. Customs and other authorities have increased the efforts to prevent illicit trafficking of nuclear material and have been furnished with modern equipment for radiation detection with the assistance of the IAEA.

In case of the detection of nuclear material, the reaction of officers depends on the classification of the detected isotope. Whether it is natural occurring nuclear material (NORM), nuclear material used for industrial applications (IND), isotopes like I-131 or Ga-67 which are in use with medical treatment (MED) or other, possibly dangerous special nuclear material (SNM).

Since the reaction, the response to an incident depends very much on the classification of the detected isotope, it is extremely important that the isotopes can be clearly identified and allocated to a library properly by the identification software on the devices.

To make the devices even faster and more reliable, a combination of the ROI method and the derivative peak-search has been developed: A quick live examination of a spectrum which is just being recorded, using the ROI method is done. As soon as the integral counts in the spectrum reach a certain limit, a derivative peak search is being started to search for further peaks. The aim of this method is to reach the highest possible detection abilities paired with the least computing- and electrical power consumption. Since the software products combining those two methods are still in development, the task has been carried out using two different software products, one for the ROI method and one for the derivative peak search.

Smugglers are, of course, also aware of the methodologies and workflows at borders and control points. That is why the issue examined in this section is very important. It is a fact, that some "medical" isotopes can be easily used to mask threatening isotopes like U-235 or Pu-239, because some lines in the gamma-spectra occur in both, the medical and the SNM isotope's spectra. This is a hardened task for the examined identification software, which should be able to clearly identify threatening isotopes even if there are large amounts of a medical isotope present.

This section will have a look at some combinations of MED and SNM isotopes and the ability of IdentPro and Identify to distinguish properly. The results should provide an overview of the strengths and failings and room for possible improvements. This applies of course for the stand-alone software products as

well as for any software to be developed using a combination of the two identification methods.

The following combinations are of high interest, not all of them have been tested though:

- **Pu-239 + I-131** (tested)
- **U-235 + Ga-67** (tested)
- **U-235 + Tl-201** (tested)
- **U-235 + Ho-166m** (tested)
- Eu-152 + Co-60
- Cs-137 backscatter peak + U-235
- Pu-239 + Ba-133
- Pu-239 + Lu-177

4.5.1. Procedure, Methodology & Setup

The following procedure has been developed to examine the abilities and disabilities of the two software products:

1. Measuring long-term spectra of the medical isotopes in the mixture (e.g.: I-131) with a CZT500 detector.
2. Scaling of those spectra with IdentPro to a total of 1000 counts and storing those downgraded spectra to disc.
3. It is a must, that the scaled spectra can be clearly identified by IdentPro!
4. Using the built-in functionality of IdentPro to add modicums of the threatening isotope which should be masked (e.g.: Pu-239).
5. The fraction of the threatening isotope is increased until IdentPro is able to identify both isotopes faultlessly (e.g.: I-131 and Pu-239)
6. The mixed spectra are stored to disc for further treatment.
7. The mixed spectra are now examined with "Identify" and the software should be able to find the gamma-lines of the threatening isotope which should not occur in the spectra of the pure medical isotope. In real life it is not really important to find the exact gamma lines of some isotopes, but to be sure that there is something in the spectra that should definitely not be there and then trigger an alarm.
8. If "Identify" is able to find peaks which must not occur in the pure isotope's spectrum, the fraction of the threatening isotope in the mixture is reduced with IdentPro until "Identify" doesn't find any additional peaks.

9. In this way it is possible to discover a fraction of the threatening isotope for each of the software-products, where the isotope can be clearly identified.
10. To be certain of those fractions the simulations have been repeated for at least ten times.

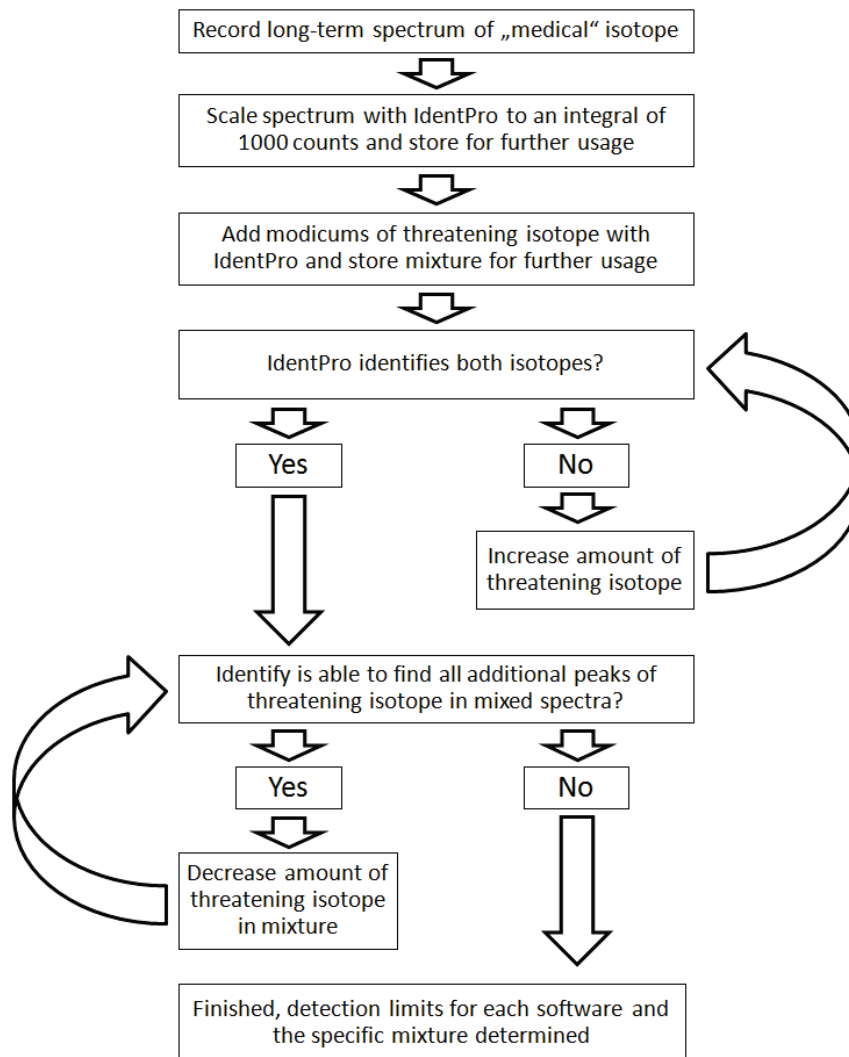


Fig. 4-14 Flow chart of the procedure to resolve the detection limits for isotope mixtures for the tested software

A comparison of the originating spectra and the mixtures is not possible, because IdentPro simulates the fractions of the threatening isotope each time one simulates a mixture! To enable a comparison of the spectra, the mixtures have

been reproduced using WinSPEC for Technicians and have then be tested and reproduced with “IdentPro” and “Identify”.

The sensitivity parameter of Identify has been set to 2,4.

The detection sensitivity parameter of IdentPro has been set to 1, the Gain was left unchanged.

4.5.2. The MED and SNM isotope libraries

As mentioned above nuclides are categorised in libraries in order to simplify the reaction to incidents by non-professional users. The MED library contains of isotopes which are commonly used for medical applications, while the SNM (special nuclear material) library contains basically the important uranium and plutonium isotopes which can be directly/through certain processes be used e.g. for nuclear bombs or dirty bombs.

The following tables show the nuclides of both libraries with some of their most important gamma-ray energies.

	Half Life	Important Energies [keV]					
F-18	109,8 m	511,00					
Cr-51	27,7 d	320,00					
Ga-67	78,3 h	91,26	93,31	184,58	208,95	300,21	393,53
Tc-99m	6,019 h	140,51					
Pd-103	17,002 d	295,95	357,46	497,07			
In-111	67,917 h	171,28	245,40	537,00			
I-123	13,3 h	159,00	346,35	440,02	505,33	528,94	538,54
I-125	60,139 d	27,47	35,49				
I-131	8,041 d	80,18	284,30	364,48	636,90	722,91	
Xe-133	5,291 d	31,00	81,00				
Ir-192	74,016 d	205,78	295,95	308,44	316,49	468,06	604,4
Tl-201	73,5 h	135,34	165,88	167,43			

Table 4-17 the MED nuclide library

	Half Life	Important Energies [keV]					
U-233	1,585E+05 a	54,69	97,21	146,35	164,60	217,97	245,35
		291,35	317,16	440,50	467,00	1566,00	
U-235	7,038E+08 a	93,35	143,76	185,72	202,12	205,31	
U-238	4,699E+09 a	13,00	63,29	92,60	112,81	766,60	1001,03
Pu-239 ¹⁶	2,413E+04 a	13,60	51,62	98,00	129,28	375,01	413,00

Table 4-18 the SNM nuclide library

It is obvious, that some of the major gamma energies in the spectrum of some SNM isotopes can be found in the spectra of some MED isotopes. This is the background of the task: Are the software products able to distinguish between two isotopes, even if some of the lines in the gamma spectra are much the same or do overlap in a significant way?

4.5.3. Results

For each medical isotope 10 samples have been produced with an integral of 1000 counts. To each of those spectra pieces of the SNM-spectra have been added and the results have been examined with IdentPro and Identify.

4.5.3.1. Pu-239 + I-131

Original spectra: PU93_scaled_0-5microSvph_run1.spe
ROI01-15min-I131-0_6-czt500-02e.spe

isotope	counts total	real-time [s]	dose-rate [μ Sv/h]	dose total [μ Sv]	cnts/s	nSv/s
Pu-239	227577	3600	0,59	0,50	74,59	0,16
I-131	30840	900	0,61	0,15	34,27	0,17

Table 4-19 Basic data of original spectra Pu-239 and I-131

The I-131 spectrum was scaled 10 times to approx. 1000 integral counts resulting in a simulated real-time of 29 seconds with an average total dose of $4,911 \pm 0,164$ nSv.

¹⁶ Plutonium is identified as either weapons grade, fuel grade, or power reactor grade depending on the percentage of P-240 that it contains. Weapons grade plutonium (WG) contains less than 7% P-240. Fuel grade plutonium contains from 7% to less than 19%, and power reactor grade (RG) contains 19% or more P-240.

spectrum name	simulated counts	total dose [nSv]
i131_29s_0.spe	965	4,772
i131_29s_1.spe	1028	5,083
i131_29s_2.spe	913	4,515
i131_29s_3.spe	1012	5,004
i131_29s_4.spe	981	4,851
i131_29s_5.spe	979	4,841
i131_29s_6.spe	1006	4,975
i131_29s_7.spe	1011	4,999
i131_29s_8.spe	1028	5,083
i131_29s_9.spe	1008	4,984
mean value	993,100	4,911
standard deviation	35,044	0,173

Table 4-20 Simulated spectra of I-131 with an average total counts of 1000

The examination of the original I-131 spectra with Identify discovers 4 gamma-lines, which are obviously not “dangerous”:

- 83,67 keV
- 282,97 keV
- 364,23 keV
- 635,75 keV

Those lines may also appear in the mixed spectra because they can be assigned to I-131.

The simulated spectra of I-131 with a real-time of 29 seconds and a integral count of about 1000 look like this in IdentPro (top) and Identify (bottom):

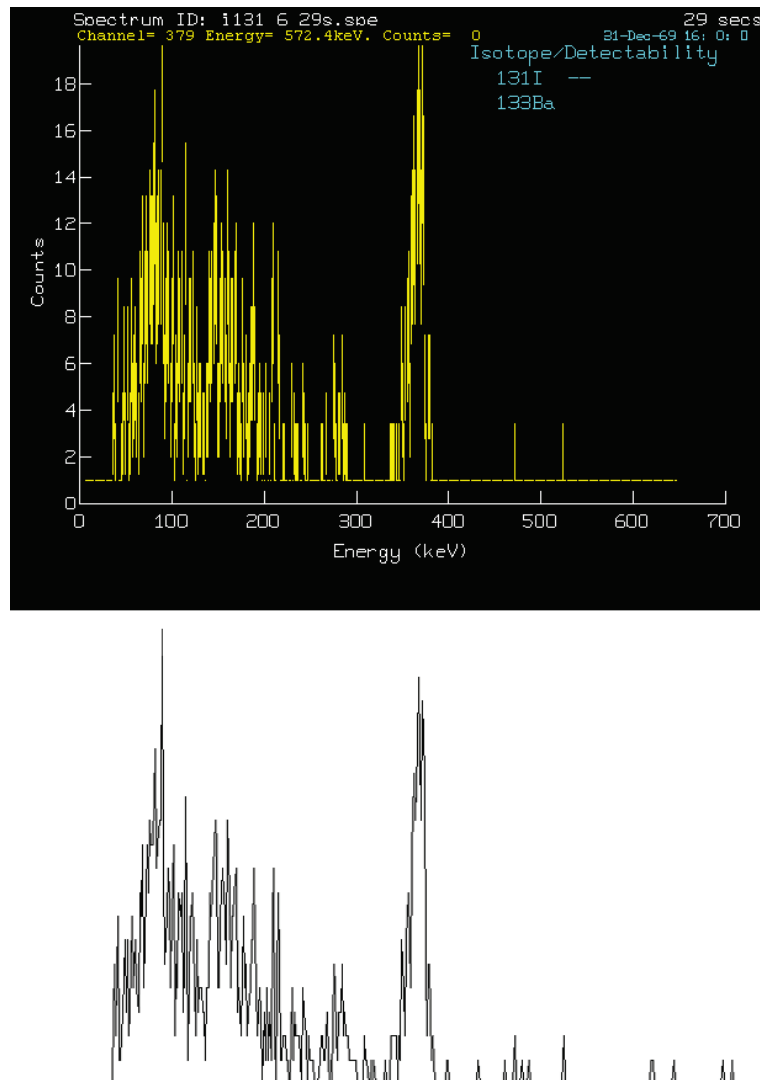


Fig. 4-15 Simulated spectrum of I-131 with a real-time of 29 seconds and a total count of 1000

Examining the 10 base-spectra IdentPro identifies I-131 as well as Ba-133 which is actually not there. Identify always discovers only one or two peaks, but is able to identify I-131 without any problems.

As a next step a small amount of the SNM-spectra, in this case Pu-239 has been added. With an amount of 1,97nSv (~12 seconds simulated real-time – “Series 1”) IdentPro is able to detect Pu-239 (weapon grade) in 80% of the cases, while Identify would trigger an alarm in only 40% of the tests.

The amount of Pu-239 was increased to 2,45nSv (~15 seconds simulated real-time - “Series 2”). IdentPro reaches 90% detection probability and Identify 80%.

Increasing the amount of plutonium to a total of 3,27nSv (~ 20s simulated real-time – “Series 3”) results in proper identification of both software products. The ration of I-131 to Pu-239 is about 15 to 10 in this case.

In the following table the results are displayed in an overview. The values indicate the gamma energy in keV; green marked values appear in the original I-131 spectrum and are not indicating any threatening nuclide present; orange marked values cannot be assigned to I-131 and should therefore result in an alarm; values in brackets are peaks which couldn't be identified by Identify with absolute certainty.

	_0	_1	_2	_3	_4	_5	_6	_7	_8	_9
Series 1				60,47						
~4,9nSv I-131	105,91		102,42	103,18					110,65	
~1,97nSv Pu-239	367,60	365,68	363,74	364,34	365,26	365,87	364,77	364,43	362,35	365,26
				[412,63]						
Series 2		[58,49]	[55,68]		65,05	63,29	[63,47]	59,67		[59,5]
~4,9nSv I-131	101,12	105,60	106,15	[98,94]	101,83	102,23	[102]		104,13	99,73
~2,45nSv Pu-239			[167,49]		159,58	[165,8]				[115,89]
		207,10								
	276,69									
	367,16	364,43	364,35	362,44	363,62	363,65	365,74	367,42	363,52	364,42
						[636,03]				
Series 3		60,40				63,09			60,32	59,44
~4,9nSv I-131	101,67	101,17	101,18	102,55	102,25	104,32	105,74	105,34	102,23	101,29
~3,27nSv Pu-239					161,05					
		287,28	286,90							285,67
	366,55	363,85	362,62	363,95	367,38	365,32	366,05	364,45	362,15	364,99
		626,02								

Table 4-21 Identified peaks with Identify - results of simulated spectra with I-131 and Pu-239

It is nice to see, that even at a lower dose rate of the threatening Pu-239, the identification algorithms of both software products are able to trigger a positive alarm in 100% of the events.

The following graph shows a sample spectrum of "Series 3" with a ratio I-131 to Pu-239 of 15 to 10.

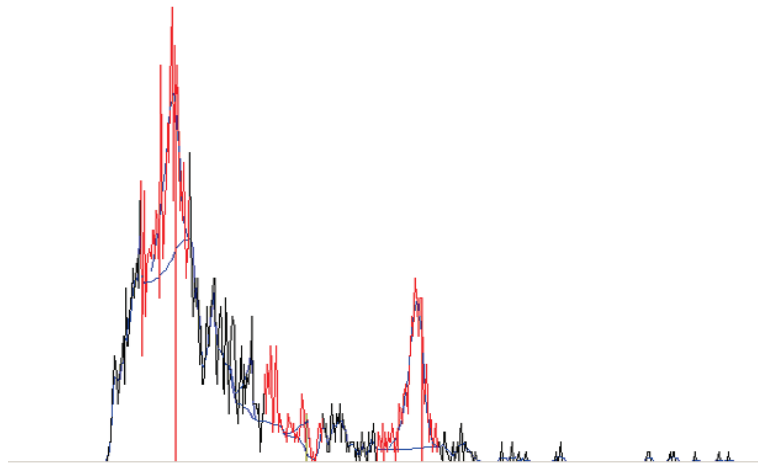
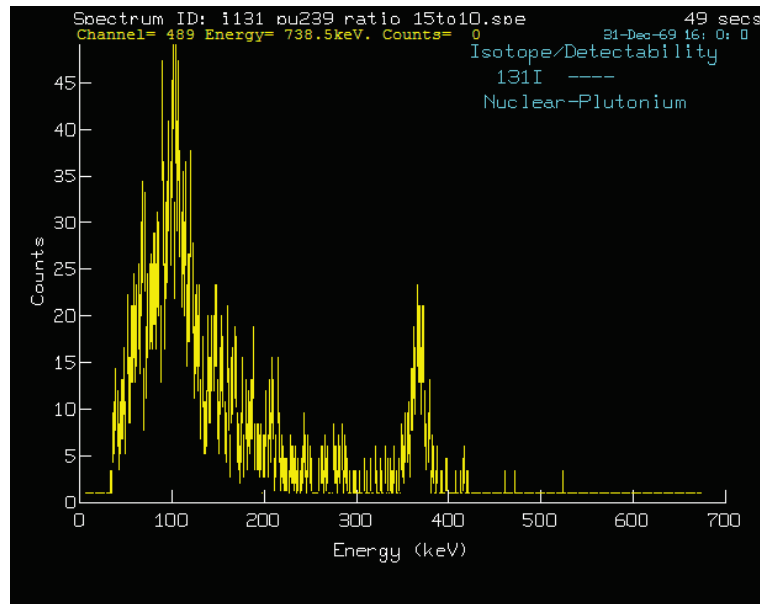


Fig. 4-16 3,27nSv Pu-239 added to 4,9nSv I-131 and examined by IdentPro (top) and Identify (bottom)

4.5.3.2. U-235 + Ga-67

Original spectra: 20min-U235-20-02c.spe
20min-Ga67-0_6-czt500-02f.spe

isotope	counts total	real-time [s]	dose-rate [$\mu\text{Sv/h}$]	dose total [μSv]	cnts/s	nSv/s
U-235	135601	1200	0,61	0,20	113,00	0,17
Ga-67	149951	1200	0,66	0,22	124,96	0,18

Table 4-22 Basic data of original spectra U-235 and Ga-67

The Ga-67 spectrum was scaled 10 times to approx. 1000 integral counts resulting in a simulated real-time of 8 seconds with an average total dose of $1,473 \pm 0,042 \text{ nSv}$.

spectrum name	simulated counts	total dose [nSv]
ga67_8s_0.spe	1027	1,506
ga67_8s_1.spe	949	1,392
ga67_8s_2.spe	1018	1,494
ga67_8s_3.spe	982	1,440
ga67_8s_4.spe	1024	1,502
ga67_8s_5.spe	995	1,460
ga67_8s_6.spe	984	1,444
ga67_8s_7.spe	1005	1,474
ga67_8s_8.spe	1050	1,541
ga67_8s_9.spe	1003	1,472
mean value	1003,700	1,473
standard deviation	28,347	0,042

Table 4-23 Simulated spectra of Ga-67 with an average total counts of 1000

The examination of the original Ga-67 spectra with Identify discovers 4 gamma-lines, which are obviously not “dangerous”:

- 95,55 keV
- 184,39 keV
- 300,13 keV

Those lines may also appear in the mixed spectra because they can be assigned to Ga-67 – the following graph shows the clear identification of Ga-67 in the plain simulated spectra with IdentPro and Identify.

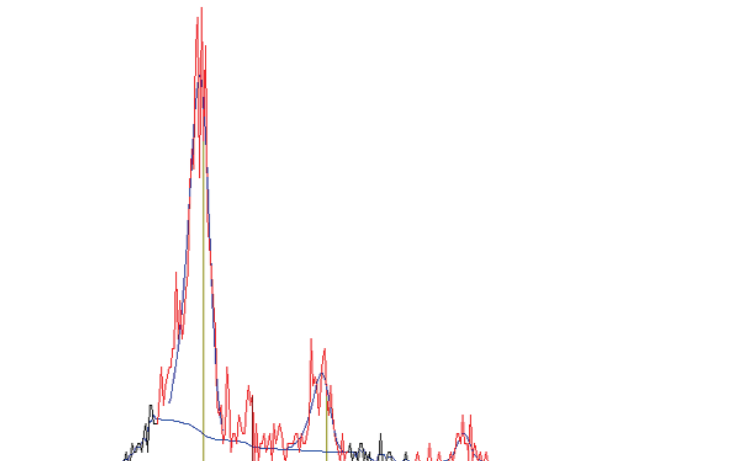
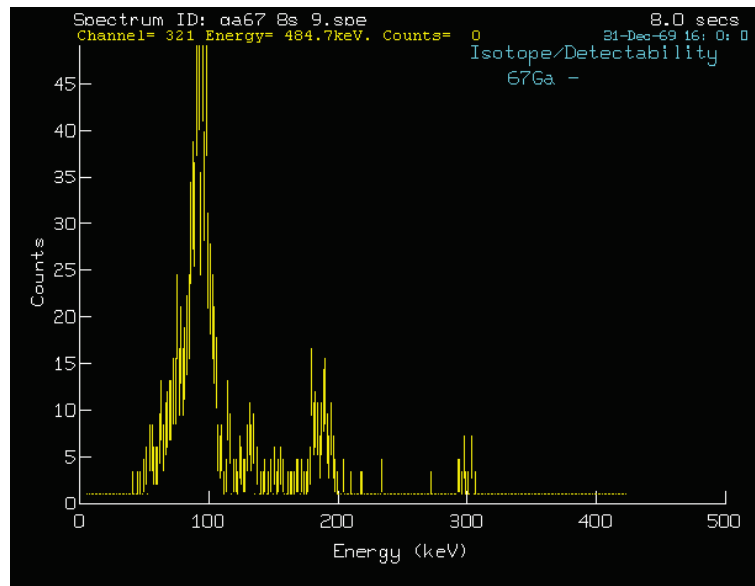


Fig. 4-17 A Ga-67 spectrum with a simulated runtime of 8 seconds, properly identified by IdentPro (top) and Identify (bottom).

In a first step a simulated spectrum of U-235 with a real-time of 10 seconds was added to the Ga-67 spectra. This equals a dose of 1,7nSv U-235 and a ration of Ga-67 to U-235 of about 10:12.

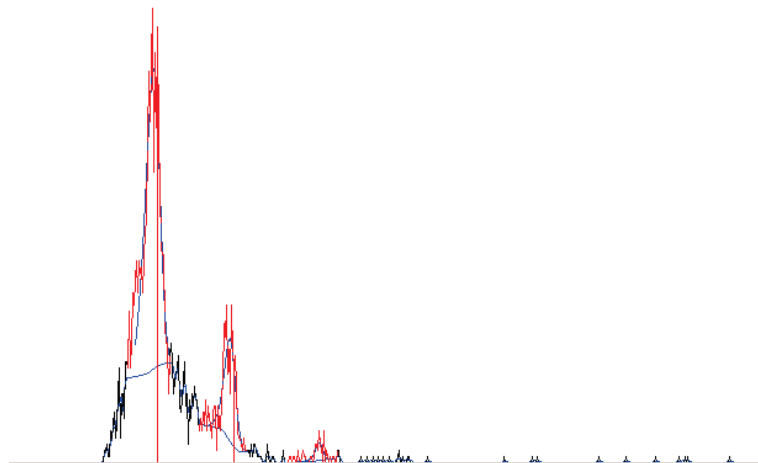
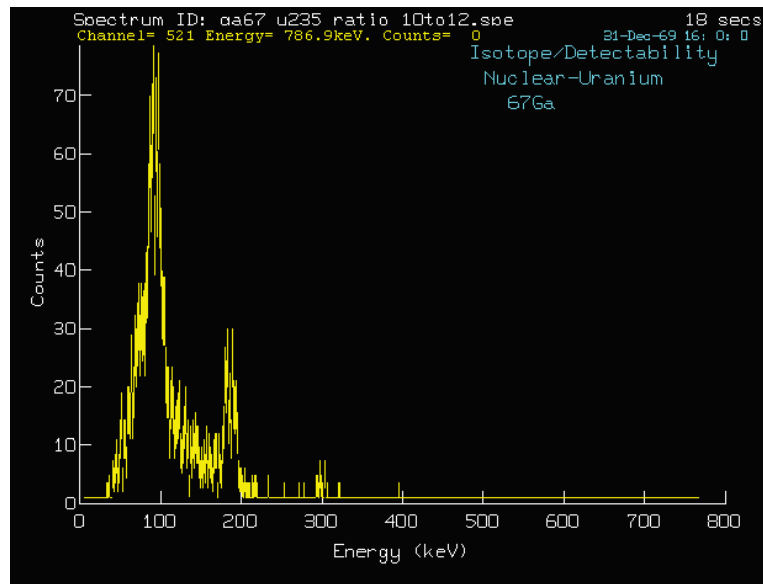


Fig. 4-18 A mixed spectrum of Ga-67 (8s) and U-235 (10s) with a total real-time of 18 seconds. Identify found some peaks, but only to few of them could be assigned to U-235

In this case, compared to the previous topic (I-131 + Pu-239), IdentPro was much faster than the peak search method of Identify. IdentPro recognised the U-235 peaks already, when the added dose of U-235 was only $\sim 1,7\text{nSv}$. Identify missed most peaks and was not able to identify U-235 in most cases. The recognised peaks were assigned to Ga-67 while the “hidden” U-235 peaks couldn’t be discovered.

As a result of this, the real-time of the added U-235 spectrum was increased further till Identify was also able to clearly recognise the U-235 behind the Ga-67 in the mixture. The following graph shows the sum of a 8 second Ga-67 spectra and a 360 second spectra of U-235 (total dose: 61nSv). The ration of Ga-67 to U-235 in this case is approx. 1:45.

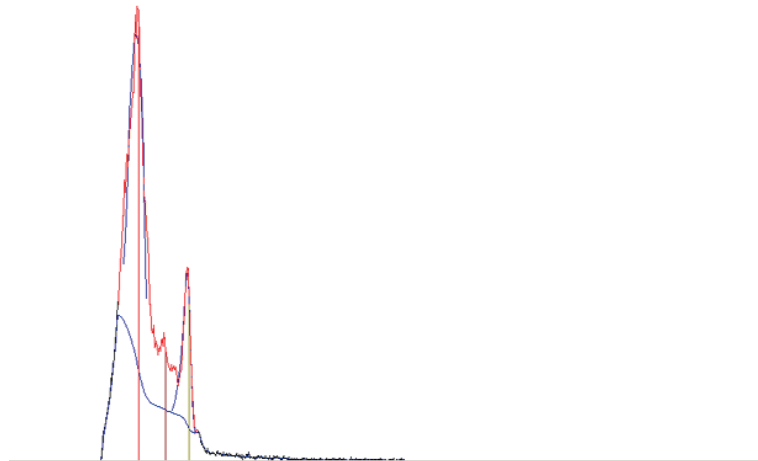
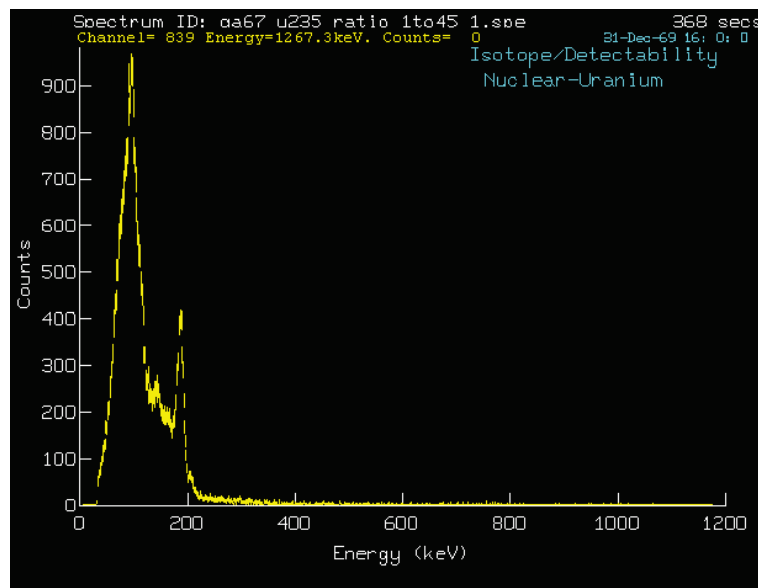


Fig. 4-19 Spectrum of a mixture of Ga-67 and U-235 in a ratio of 1:45. The amount of U-235 overwhelms the Ga-67, so that IdentPro doesn't recognize the MED isotope any more.

Identify manages to recognise peaks in 100% of the cases which cannot be assigned to Ga-67 and must therefore lead to an alarm. The results of “Series 6” are displayed in the following table.

	_0	_1	_2	_3	_4	_5	_6	_7	_8	_9
Series 6										
~1,47nSv Ga-67		72,93							73,51	
~61nSv U-235		85,01					84,94			
	96,66	96,71	96,64	96,68	95,7	96,71	96,81	95,74	96,81	96,76
		109,96				110,34			111,24	111,06
	144,42	144,04	143,42	144,17	144,37		143,14	144,45		
	186,98	187,14	186,9	186,91	187,12	186,91	187,04	187,04	187,11	186,05
						295,33			299,86	
									1000,77	

Table 4-24 Overview of peaks recognized by Identify within the sample spectra containing of 1,47nSv Ga-67 and 61nSv U-235.

In this case the abilities of the derivative peak-search stay way behind the ROI method of Ray Gunnink’s IdentPro. Since a ratio of 1:45 of the MED isotope comparing to the SNM is not likely and – in a realistic scenario – the smuggler would have succeeded, this experiment indicates the need of a combination of the two identification methods.

To confirm this intent, the same experiment has been carried out with Tl-201 as MED isotope.

4.5.3.3. U-235 + Tl-201

Original spectra: 20min-U235-20-02c.spe
20min-Tl201-0_6-czt500-02f.spe

isotope	counts total	real-time [s]	dose-rate [$\mu\text{Sv/h}$]	dose total [μSv]	cnts/s	nSv/s
U-235	135601	1200	0,61	0,20	113,00	0,17
Tl-201	267447	1200	0,54	0,18	222,87	0,15

Table 4-25 Basic data of original spectra U-235 and Tl-201

The Tl-210 spectrum was scaled 10 times to approx. 1000 integral counts resulting in a simulated real-time of 5 seconds with an average total dose of $0,761 \pm 0,017 \text{ nSv}$.

spectrum name	simulated counts	total dose [nSv]
tl201_5s_0.spe	1181	0,795
tl201_5s_1.spe	1105	0,744
tl201_5s_2.spe	1123	0,756
tl201_5s_3.spe	1089	0,733
tl201_5s_4.spe	1151	0,775
tl201_5s_5.spe	1138	0,766
tl201_5s_6.spe	1126	0,758
tl201_5s_7.spe	1145	0,771
tl201_5s_8.spe	1111	0,748
tl201_5s_9.spe	1132	0,762
mean value	1130,100	0,761
standard deviation	25,963	0,017

Table 4-26 Simulated spectra of Tl-201 with an average total counts of 1000

Identify detects 4 peaks in the plain Tl-201 spectrum.

- 71,69 keV
- 79,93 keV
- 136,84 keV
- 167,78 keV

Those gamma-lines may appear in the examined mixtures as well as they can be clearly assigned to Tl-201.

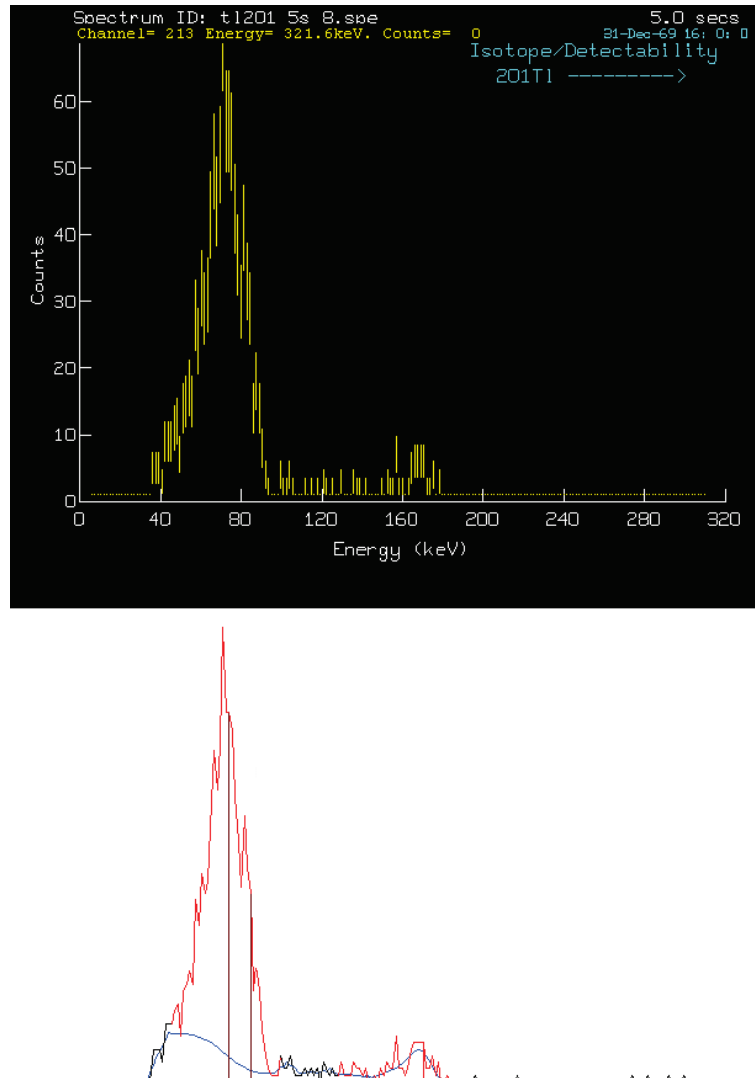


Fig. 4-20 Sample spectrum of Tl-201 with a simulated real-time of 5 seconds. On the top we see IdentPro identifying Tl-201 with absolute certainty and at the bottom Identify presents the detected peaks.

Adding small amount of U-235 did not lead to a proper identification of U-235 behind Tl-201. Increasing the real-time to about 10 seconds (1,7nSv U-235) would result in Identify triggering a red alarm in 50% of the examined cases. The dose of U-235 was therefore raised to 2,37nSv which corresponds to a simulated real-time of 14 seconds. The resulting spectrum with a total real-time of 19 seconds can be clearly identified as a mixture of Tl-201 and U-235 in 100% of the

cases. Identify definitely discovers a peak at 188keV which is definitely not a part of the Tl-201 spectrum.

The ration of Tl-201 to U-235 at this stage is about 1:3.

IdentPro detects the nuclear uranium in 100% of the cases, even at low ratios of 1:1,5 and 1:2. The following figure shows a sample spectrum with a ration of 1:3 in IdentPro and Identify.

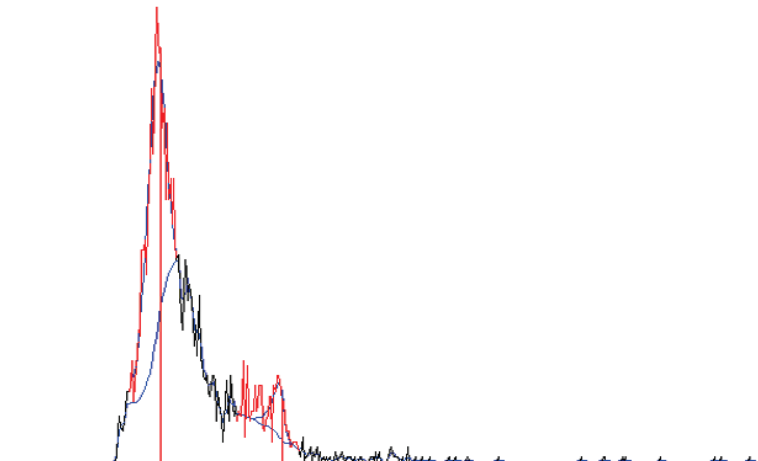
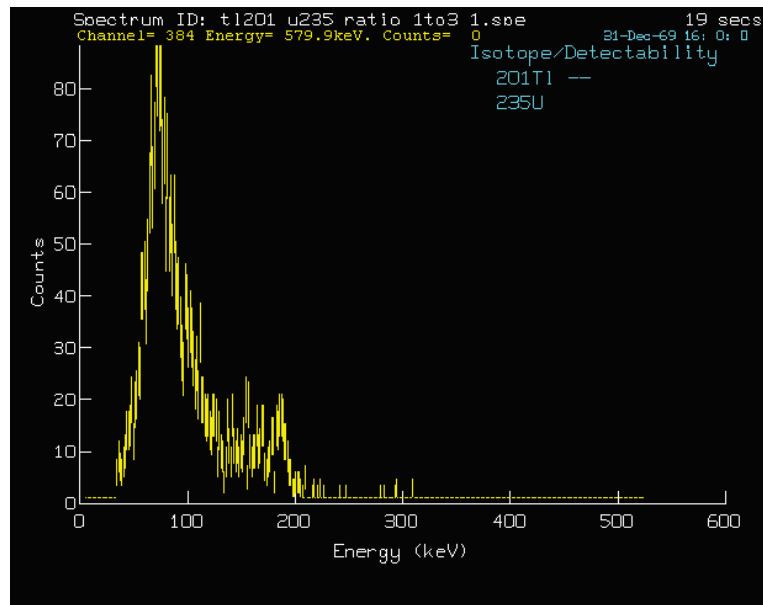


Fig. 4-21 Sample spectrum of Tl-201 mixed with U-235 at a ratio of 1:3. IdentPro is able to identify U-235 next to Tl-201 and Identify discovers an additional peak at 188keV.

	_0	_1	_2	_3	_4	_5	_6	_7	_8	_9
Series 1	72,79	73,37	73,3	73,96	75,37	72,74	73,86	75,96	74,75	72,92
~0,76nSv Tl-201					[111,77]					
~1,7nSv U-235		[168,59]								
	186,34	[187,4]	[188]	187,56	187,77	185,57		187,9	[186,4]	[189,56]
Series 2	74,67	73,44	74,34	73,73	74,14	73,5	71,11	73,77	73,75	75,09
~0,76nSv Tl-201							81,52			
~2,37nSv U-235	188,08	186,72	187,52	187,03	188,59	188,14	188,43	188,27	187,9	188,17
Series 3	73,03	73,65	73,15	74,11	74,76	73,24	72,64	72,46	74,84	72,39
~0,76nSv Tl-201	96,18		97,56		[94,65]	[97,78]	96,23		96,35	
~5,93nSv U-235									144,26	
	187,7	187,85	187,69	186,52	186,81	186,95	187,1	187,02	186,87	185,52

Table 4-27 Identified peaks of 3 simulation series with Identify. Series 1 lead to an identification rate of 50%, with series 2 it was possible to identify the additional peak in every case. Series 3 was a simulation with a ratio of about 1:7 to see whether Identify could find even more additional peaks.

In this experiment, other than the one with U-235 and Ga-67, both software products showed very much the same capabilities to discover a hidden SNM material on a MED background. It has to be said, that the peak-search algorithm of Identify seems to have problems in some cases with a masking MED isotopes while Identify using the advanced ROI method is very fast and reliable detecting “hidden” isotopes.

As a last example the medical isotope Ho-166 has been used together with another source spectrum of U-235 (dose-rate at recording: 0,5 μ Sv/h instead of 0,61 μ Sv/h). The results are presented below in a brief overview.

4.5.3.4. U-235 + Ho-166

Original spectra: ROI01-20min-U235-czt500-02.spe
ROI01-1h-Ho166m-czt500-02b.spe

isotope	counts total	real-time [s]	dose-rate [$\mu\text{Sv/h}$]	dose total [μSv]	cnts/s	nSv/s
U-235	103064	1200	0,5	0,17	85,89	0,14
Ho-166	135711	3600	0,5	0,50	37,70	0,14

Table 4-28 Basic data of original spectra U-235 and Ho-166

The Ho-166 spectrum was scaled 10 times to approx. 1000 integral counts resulting in a simulated real-time of 26 seconds with an average total dose of $3,632 \pm 0,116 \text{ nSv}$.

spectrum name	simulated counts	total dose [nSv]
ho166_26s_0.spe	1032	3,802
ho166_26s_1.spe	974	3,589
ho166_26s_2.spe	971	3,577
ho166_26s_3.spe	990	3,647
ho166_26s_4.spe	972	3,581
ho166_26s_5.spe	1029	3,791
ho166_26s_6.spe	985	3,629
ho166_26s_7.spe	994	3,662
ho166_26s_8.spe	921	3,393
ho166_26s_9.spe	991	3,651
mean value	985,900	3,632
standard deviation	31,377	0,116

Table 4-29 Simulated spectra of Ho-166 with an average total counts of 1000

Both IdentPro and Identify were able to find all additional peaks at ratios of 1:1,2 and above. IdentPro was a little bit faster again and detected the nuclear uranium even at ratios of 10:9 (Ho-166 to U-235).

4.5.4. Conclusion

Having all the accumulated results and the dozens of spectra in mind, the advanced ROI method of Ray Gunnink within the software IdentPro seems to be the more reliable one when dealing with the masking of threatening isotopes

through MED isotopes. Even though the parameters of Identify have been very well tuned and the setup to the right detector type and the calibration has been finically accurate, the results were not comparable to those of IdentPro. Since the advanced ROI method takes advantage of its interference coefficients to compare peak heights in the spectra and relations of the peak to each other, the “plain” derivative peak search is at a slight disadvantage. However, depending on the type of mixture of MED and SNM isotopes, both algorithms have their advantages and should be further developed side by side as well as together.

As a conclusion it has to be said that a combination of both methods in one software-product would definitely make much sense and could verifiable improve the possibility of a proper identification of unknown materials in the field.

A light weight program code would not need the computing power of a personal computer, but could cope with the resources of an up-to-date PDA device. This would as well lead to further possibilities concerning the size of the read-out and identification electronics of a detector. On the other hand a PDA as computing device within a detector system could provide intrinsic gateways to centralised databases or storage systems/ -centres (cellular network, high speed internet connection, blue-tooth,...) from almost every place round the globe.

5. Conclusion and discussion of results

First of all it has to be said, that the testing and measurements of the devices as well as the software have been carried out over a period of 4 years due to several reasons. However, the results are valid because the main characteristics of the examined hardware and software products have not changed ever since. There have of course been lots of improvements on certain issues like the Graphical User Interface (GUI) or the connectivity with some devices, because the IAEA is constantly working on this and trying to push developments. The IAEA is in tight contact with manufacturing companies to assess the performance and to systematically improve the devices. Those efforts have yield fruit in the meantime but cannot be incorporated to the full extent into the state of 4 years ago described in this work.

In addition to the described tests and measurements, several other devices have been tested in the meanwhile as well - for example the Thermo Interceptor and different Backpack detector systems.

5.1. Hardware

The examined detection systems are chosen from possibly 10 or more systems the IAEA evaluates and distributes to its member states for border security, police work and other operations in the context of issues concerning nuclear material. All those devices have to match the “Technical and Functional Specifications for Border Monitoring Equipment” as stated in the IAEA Nuclear Security Series No. 1. Therefore the results for the tested devices can be somehow applied to all other handheld RIDs¹⁷, even though the strengths and deficiencies will certainly vary depending on the specific device.

¹⁷ RID: Radionuclide Identifying Device

It is difficult to describe an overall impression concerning all handheld RIDs and SPRDs¹⁸ in use by the IAEA. Since the manufacturers and their approach to solve certain problems vary, most of the comments later in this text may apply to only one or some of the devices. In some cases it is not clear to me why certain technical and functional specifications stated by the IAEA are not fulfilled or only partly executed, because the IAEA puts a lot of effort into development of those specifications and guidelines.

However – it would go beyond the scope of this work to discuss all of the issues, so only some of them are mentioned in short as follows:

- In some cases the manufacturers could possibly put more effort into the development of the Graphical User Interface (GUI). It is of course a hard challenge to strike a balance between a high tech instrument with all its possibilities and functions and the non-professional user who has to deal with this instrument in “easy mode”¹⁹, maybe even without understanding the technical background.
- Another point is the “connectivity” issue. Nowadays there are standards like USB or Bluetooth which are not “state-of-the-art”, but more like common techniques. Most of the manufacturers already provide those interfaces, but there are still some devices which are equipped with a RS-232 interface only (serial port).
- An associated topic is the provision of firmware and drivers for the devices suitable for the actual versions of the particular operating systems. That is why it is sometimes a challenge to simply connect a device to a personal computer and transfer e.g. spectra or configuration data.
- The configuration of the devices can – in some cases – be handled via computer software installed on a PC. In conjunction with the preceding topic those software products are not working well on e.g. all versions of Microsoft Windows (2000, XP, Vista,...)
- Some technical characteristics of the devices should be communicated to the end-user. Such as the “minimum integral count” particularity of the ICX IdentifINDER. If the integral counts in a spectrum are too low, the device does not trigger the identification process. This is obvious

¹⁸ SPRD: Spectral Personal Radiation Detector

¹⁹ All devices have to be equipped with an „easy“ and an „expert“ mode. The simple mode is intended for the use by non-professional users in terms of radiation detection, e.g. policemen or fire brigade and provides restricted functionality but important tools like the “search mode” and the “identification mode”. The professional mode offers more functionality as calibration, examination of spectra and so forth.

and understandable to a professional, because it is necessary to rely on certain statistics to apply the template matching method of ICx, but on the other hand it might confuse a non-professional user who is not aware of the technical background.

- The IAEA's specifications require the output of spectral data in plain ASCII format (plain text files) and this – of course – results in a variety of different output formats, depending on manufacturer and device. In terms of a quick and uncomplicated readout, processing and sharing/reporting of spectral data this should be brought to a standard. There are intentions of the IAEA to push such a standard in form of a XML²⁰-style marked-up output file. In the US there are also efforts to establish such a standard. In near future it should be possible that most of the devices understand the XML-standard and are able to generate this output as well. An example of a possible output file is presented in Appendix C.

Concluding the statements on the hardware I state the following:

- Because of the small dimensions of the devices and the small built-in crystal it is obvious that the detection capabilities of those class of devices are naturally limited. Even though the devices have to recognise certain amounts of Pu-239 as well as U-235 behind a 10mm Pb shielding (IAEA specifications), it is a hard task to identify a SNM isotope behind a large background of MED or NORM material.
- In addition a neutron search device (3He tube), specially developed for the IAEA, can be used to find neutrons, emitted by fissile material like Pu isotopes, even if the gamma rays are absorbed.
- The IAEA has modified the specifications for RID's in that way, that a higher weight of the devices is acceptable. As a consequence it is possible to integrate larger crystals, such as for example a 0,74" x 4" NaI crystal in ICx's radHUNTER and other new devices from various manufacturers.
- However it is important to know about the capabilities of the devices in use. Handheld RIDs are intended to indicate a need for action and induce doubt. They are used to discover, that something exceeds the normal radiation level and that there is a need to take further measures.

²⁰ XML: Extensible Markup Language

5.2. Software

The issue of the standardised spectrum output format obviously affects the software products as well. Most of the software is able to read different spectral formats but it would definitely make sense to get a standard working. It would increase the possibilities to share spectra between experts and possibly automate the process of incident reporting and identification of nuclides.

As mentioned before a combination of more than one methods to examine spectra would definitely make sense. In the first place it would be better not to rely on a single method and in the second place, some methods show deficiencies which could be compensated with a second method.

For example the ROI method works very well if the ROIs and the corresponding interference coefficients are well known and stored on the devices taking the measurements. But what if the ROI for a certain peak (gamma line) of a threatening isotope is not included in the ROI-set? In that case the ROI method would not be able to “see” this peak and identify the emitting isotope. The detection would fail.

The situation is very similar to the template matching method. Due to relatively poor computing power of handheld devices and small memory to store sample templates, the hit-rate of this method is limited. For example the ICx IdentIFINDER uses only fixed templates in combination with the ability to interpolate between two templates to fit the requirements. A new method called “Gadras” improves the template matching method. Used together with a well defined and calibrated detector the method takes account of the surrounding environment and starts a quick monte carlo simulation based on the available data (source term, measurements and the environment). The simulation is then compared to the measurement to guarantee a reliable result.

In my opinion - assuming a perfect world - the IAEA runs a centralized application with an attached database where public authorities of member states as well as research institutions and manufacturers of radiation detection devices have access.

If an incident happened on some border crossing, the customs officer would record sample spectra and transfer them to the centralised IAEA application. Either directly using the cellular network, or via a personal computer, PDA or cellular phone which could be connected to the device through Bluetooth or a wireless local area network (Wi-Fi).

The application would immediately start the identification process, using various available techniques and algorithms to find peaks and identify nuclides.

The algorithm could also rely on a huge database of stored samples and templates from preceding incidents or scientific work and could therefore provide well-founded results.

When the identification process finishes, the officer at the border crossing would receive the results through the same channel as he sent it to the centralised application and can then take necessary measures.

This could of course be extended; the centralised application could as well trigger an alarm, when a spectrum of some incident could not be identified or if some SNM was detected. The system could as well be used by IAEA inspectors, universities or other research facilities and - of course - the manufacturers.

Appendix A: Nuclide libraries

Each manufacturer of RIDs as well as programmers of software products use certain nuclide libraries, where each isotope is related to peaks which should occur accordingly in the isotope's spectra. The following table is an aggregated summary of some nuclide libraries which were used for this work. It is interesting to see, that the stated energies for some peaks vary in a sometimes wide field. Some libraries rely on only few peaks, while others contain almost every gamma-peak. All energies are denoted in keV.

library	Polimaster polismart	Atomtex	SpecMon watchmon	Target customs	Aspect mkc mng.	Aspect mkc a03	IdentPro	Canberra Insp. 1000	ANSI 42.34
isotope									
Am 241		26,000 60,000 120,000	59,530 98,940 102,950	26,350 62,000	59,537	59,500		59,541 98,930 102,930 122,990	59,540
Ba 133	355,990	31,000 53,000 81,000 118,000 157,000 292,000 356,000 455,000	80,980 276,380 302,850 383,840	53,160 80,990 160,600 276,400 302,850 356,010 383,850		290,000 296,000 296,000 358,000 609,000	302,000 357,000 383,700	80,997 276,400 302,851 356,005 383,848	80,997 356,005
Bi 207				72,800 74,970 84,800					

	569,150		569,150	87,300 569,620 897,700 1063,650 1442,200 1770,180	569,700	569,700	569,600		
	1063,090		1063,090		1063,660	1063,700	1063,600		
Cd 109				22,100 25,000 87,700				88,032	
Ce 139	165,850							165,850	
Co 57				6,400 7,060 14,410					
		33,000 85,000							
	122,060	122,000	122,060	122,060 136,480		122,100	122,050 136,500	122,063 136,476	122,063 136,476
		252,000 695,000							
Co 60		78,000 221,000 910,000							
	1173,230 1332,500	1173,000 1333,000	1173,230 1332,500	1173,230 1332,510	1173,240 1332,500	1173,200 1332,500	1173,230 1332,510	1173,216 1332,486	1173,216 1332,486
Cr 51		212,000 320,000							
	320,080	320,000	320,080				320,000	320,082	
Cs 134				475,350 563,330 569,370 604,740 795,800 801,860 1038,500 1167,860 1365,130					
Cs 137		32,000 78,000 186,000 203,000		32,000				32,194 185,700	

	661,660	442,000 662,000 1327,000	661,660	661,640	661,660	661,700	661,650	661,650	661,650
Eu 152						10,000			
		40,000 78,000 84,000							
	121,770	122,000	121,770	121,780		121,800	121,800	121,782	
		173,000							
	244,690	245,000	244,690	244,700		244,700	244,700	244,697	
	344,280	344,000	344,280	344,270		344,300	344,300	344,279	
		444,000		444,000		444,000			
		566,000				571,000			
						676,000			
	778,910	778,000	778,910	778,900		778,900		778,904	
				867,400				867,373	
	964,110	964,000	964,110	964,000		964,100		964,079	
				1086,400				1085,869	
	1100,000	1103,000				1102,000			
				1112,050				112,069	
	1408,000	1408,000	1408,000	1407,920		1408,000		1408,006	
F 18			511,000				511,100		
Ga 67				8,630 9,570					
		75,000 90,000		91,270 93,310	93,311			93,100	93,311
		140,000							
	184,570	186,000	184,570	184,580	184,569	184,600	184,590	184,577	184,577
	208,950		208,950	208,950					
	300,210	300,000	300,210	300,220	300,219	300,200	300,020	300,219	300,219
						358,000			
	393,520	393,000	393,520	393,530			393,650	393,529	
		495,000							
		794,000 888,000				609,000			
Hg 203								279,190	
I 123		27,000 78,000							
	158,980	159,000	158,980	159,000			159,000	159,100	158,970

	645,160 1479,220								
K 40	1460,830	1144,000 1461,000	1460,830	1460,750	1460,830	1460,800	1460,000	1460,810	1460,810
Mn 54	834,820	78,000 202,000 596,000 835,000		5,410 5,950 834,810					
Na 22	511,000 1274,520	78,000 177,000 312,000 511,000 1006,000 1274,000	511,000 1274,520	511,000 1274,550			511,000 1274,500	1274,530	
Np 237	312,010 340,580	87,000 312,000	86,520 98,430 111,020 312,010 340,580				311,900 405,000	86,490 312,170 340,810	86,477 143,249
Pd 103	357,450 497,050		39,750 62,500 294,950 357,450 497,050	39,740	357,450 497,080	295,200 357,500 497,100	21,000 295,000 357,450 497,080	39,748 62,410 294,980 357,450 497,080	
Pu 239		22,000 50,000 78,000 96,000 128,000 200,000 330,000	51,620 129,280 203,550 332,800	38,690 51,620 98,440 129,270 332,840		214,00 RG 210,00 WG 334,00 RG	51,630 98,800 129,296 203,500 208,000 332,000	98,780 129,294	

	340,000 375,010		344,940 375,010		348,000	343,00 WG		345,000 375,054	
		380,000 390,000				382,00 RG 382,00 WG	376,000		
	413,690		413,690 413,710 451,440		648,000		413,710	413,713	413,712
						658,00 RG 662,00 WG 918,00 RG			
Pu 241	208,000 332,360								164,610 208,000
Ra 226		47,000 77,000 145,000							
		186,000	186,110	186,210	186,000	186,000		186,211	186,100
		242,000	241,910				241,960		
		295,000	295,090		295,200	295,200	295,200		295,220
	351,860	352,000	351,860		351,925	351,900	351,920		351,990
	609,310	609,000	609,310		609,000	609,000	609,270		609,312
		777,000 934,000							
	1120,270	1120,000	1120,270						1120,287
		1416,000							
	1764,490 2204,090		1764,490 2204,090						1764,494
Se 75		97,000						96,734	
	121,110		121,110					121,116	
	136,000	133,000	136,000				136,000	136,000	
		199,000						198,606	
	264,650	269,000	264,650				264,600	264,658	
	279,530		279,530					279,542	
	400,650	401,000	400,650				400,600	400,657	
Sm 153			41,540 69,670 103,180 172,850 531,430 596,720						

Sn 113	391,680							391,688	
Sr 85								513,990	
Tc 99m		78,000 90,000 140,470	140,000	140,470				140,510 140,511	140,508
Th 228		43,000 77,000 125,000 238,570 303,000 384,000 503,000 583,020 729,000 862,000 1626,000 2614,350		238,570 240,760 277,280 300,030 510,600 583,020 727,240 860,300 2614,350					
Th 232		61,000 77,000 125,000 238,570 324,000 334,000 384,000 503,000 583,020 738,000 911,150 918,000 968,970 1592,350 2103,350 2614,350		238,630 583,140 911,070 968,940 2614,600	238,632 583,191 920,000	238,600 583,200 918,000	238,630 583,140 911,204 918,000 968,971 1588,200 1620,500 2614,533	77,110 238,632 338,320 583,191 727,180 911,070 968,971 1588,200 1620,500 2614,530	238,322 338,322 583,190 911,070 968,971 2614,530
Ti 44	511,000 1157,000 1499,450								

TI 201		43,000		68,890 70,820 80,200 82,500	70,000	70,000		69,000 81,300	
		77,000							
		128,000	135,270	135,300				135,340 165,880	135,340
		167,000	167,400	167,400	167,400	167,400	167,430	167,430	167,430
TI 204								68,895 70,819 80,300	
U 233				42,440 54,690				42,400 54,690	
		78,000							
		93,000		97,140				97,210	
		145,000		146,340 164,540				146,345 164,600	
	193,590		193,590						
	217,970	217,000	217,970						
		313,000		291,340 317,150				291,354 317,160	291,354
	440,340		440,340		440,500	440,500	441,000		
	467,000		467,000						
								583,000 662,000	
	1566,000		1566,000		1567,000	1567,000			
U 235		63,000							
		78,000							
		93,000				94,000			
		147,000	143,780	109,120 143,780				143,760	143,760
			163,370	163,360				163,330	
	185,720	186,000	185,730	185,720	186,000	186,000	185,720	185,715	185,715
				194,910					
				202,130				202,110	
	205,310		205,330	205,310				205,311	
								609,000 661,700 918,000	
U 238		63,000	63,290					63,290	63,290

		78,000 93,000 186,000						92,590	92,600
			258,000		186,000				
	766,350 1001,020	766,000 1001,000	766,350 1001,020		766,370	609,000 766,400 1001,900	1001,000	766,600 1001,030	1001,030
Xe 133		31,000 82,000	80,980				81,000	80,997	80,997
Xe 135				249,650 608,600					
Y 88	898,060 1836,070				898,000 1836,000	898,000 1836,000		898,021 1836,010	
Zn 65	511,000 1115,510								

Table 5-1 Comparison of different nuclide libraries

Appendix B: IdentiFINDER Batch File

This batch file was used to repeat the verification measurements with Target IdentiFINDER in Part 4.4

```
# initialise repeat value
SetPar RepeatVal 0

# set marker for loop
:LOOP

# spectrum clear
SendNANOSString 1 "spc"

# set preset 100 sec on the real time of the device
SendNANOSString 1 "!pr 100"

# start measurement
SendNANOSString 1 "adcon"

# wait one sec more than the preset time
Incpar RepeatVal 1

# analyze spectrum
StrCat RepeatLabel "# measurement No: " $RepeatVal
SendNANOSString 1 $RepeatLabel
SendNANOSString 1 "ana"

# wait one moment
```

Wait 1

```
    # if the number of repetitions is not yet reached, jump to LOOP marker
if $RepeatVal<20
Goto LOOP
```

```
    # if finished, reset device
SendNANOSString 1 "!pr 0"
```

```
    # beep when finished
SendNANOSString 1 "deb 48"
Wait 1
SendNANOSString 1 "deb 48"
```

Appendix C: Example of a XML-based output file²¹

This example shows a possible output file with dummy-data.
Blue colour shows the Markup-Language, while black indicates data.

```

<N42InstrumentData>
  <InstrumentInformation>
    <InstrumentType>Spectrometer</InstrumentType>
    <Manufacturer>Spectrometers R Us</Manufacturer>
    <InstrumentModel>model #1</InstrumentModel>
    <InstrumentID>serial #1</InstrumentID>
  </InstrumentInformation>
  <Measurement>
    <Spectrum>
      <StartTime>2003-11-22T23:45:19</StartTime>
      <RealTime>PT60S</RealTime>
      <LiveTime>PT59.61S</LiveTime>
      <Calibration Type="Energy" EnergyUnits="keV">
        <Equation Model="Polynomial">
          <Coefficients>-21.84                12.105214
          0.006553</Coefficients>
        </Equation>
      </Calibration>
    <ChannelData>
      0 0 0 22 421 847 1295 1982 2127 2222 2302 2276
    </ChannelData>
  </Measurement>
</N42InstrumentData>

```

²¹ Source: <http://physics.nist.gov/Divisions/Div846/Gp4/ANSIN4242/xml.html>

2234 1921 1939 1715 1586 1469 1296 1178 117 107 928
760
679 641 542 529 443 423 397 393 322 272 294 227
216 224 208 191 189 163 167 173 150 137 136 129
150 142 160 159 140 103 90 82 83 85 67 76
73 84 63 74 70 69 76 61 49 61 63 65
58 62 48 75 56 61 46 56 43 37 55 47
50 40 38 54 43 41 45 51 32 35 29 33
40 44 33 35 20 26 27 17 19 20 16 19
18 19 18 20 17 45 55 70 62 59 32 30
21 23 10 9 5 13 11 11 6 7 7 9
11 4 8 8 14 14 11 9 13 5 5 6
10 9 3 4 3 7 5 5 4 5 3 6
5 0 5 6 3 1 4 4 3 10 11 4
1 4 2 11 9 6 3 5 5 1 4 2
6 6 2 3 0 2 2 2 2 0 1 3
1 1 2 3 2 4 5 2 6 4 1 0
3 1 2 1 1 0 1 0 0 2 0 1
0 0 0 1 0 0 0 0 0 0 0 2
0 0 0 1 0 1 0 0 2 1 0 0
0 0 1 3 0 0 0 1 0 1 0 0
0 0 0 0

</ChannelData>

</Spectrum>

</Measurement>

</N42InstrumentData>

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