Hardware and Software Applications in Nuclear Spectrometry

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Abstract

An overview of the status and function of current spectrometry hardware, detectors having energy-dispersive (spectrometric) properties as well as the latest developments in quantitative spectrometry software is presented.

Introduction to nuclear spectrometry

Nuclear spectrometry is basically concerned with the measurement of the energy of charged or neutral fragments or photons from nuclear decay or nuclear reactions. In this very general description, the variety of measured charged particles ranges in terms of mass from electrons to fission fragments, whereas the masses of uncharged radiation are 1 amu or less, the heaviest emitted particle being a neutron. Energies of radiation or particles go from almost zero keV to the highest energies available in nuclear reactions. We will confine ourselves in the following to the description of nuclear spectrometry of photons and alpha particles originating from nuclear decay. These two areas constitute the most important applications of nuclear spectrometry. Many details and principles presented in this paper, however, apply in the same way to the measurement of other fragments from nuclear reactions, irrespective of their origin or energy, and of course also for high-energy nuclear reaction studies.

Spectrometry of β-particles, both negatrons and positrons, especially when measured in coincidence with γ-rays is an indispensable tool for every kind of nuclear structure studies, and spectrometry of neutrons is an important tool in the analysis of high-energy nuclear reactions. However, neutron and electron spectra are normally continuous in nature and one interprets only the average or the maximum (endpoint) energy from the spectrum. Thus, most spectrometry details mentioned in the software section below will not apply to the analysis of continuous spectra, whereas hardware-related details may well be applicable.

The most important application of nuclear spectrometry is to detect and quantify radioactive nuclides in a sample, i.e. nuclear spectrometry is an analytical technique for radioactive nuclides. A strong advantage of nuclear spectrometry is its unambiguous definition of the emitting nuclide, i.e. the emitter is characterized in terms of element (Z) and isotopic mass (A). When one or several easily measurable nuclides are to be surveyed, spectrometry is also suited for status- or process-control. Nevertheless, in many cases of technical surveillance other methods of nuclide quantification are necessary. The most prominent example for the employment of an alternative method is the use of mass spectrometry instead of nuclear spectrometry in all cases where isotopic concentrations in separation facilities or very small element concentrations are to be determined. For quantitative determination of the element uranium, for example, one may use methods of colorimetry, voltammetry, fluorimetry or mass spectrometry.

There are other restrictions to nuclear spectrometry which generally hamper its application. When a nuclide has very long half-life, one will measure low count-rates and the statistical significance of data is poor. One can overcome this disadvantage by extending the counting time; however, there are practical limitations and very long-lived nuclides cannot be measured by nuclear spectrometry. The same argument applies for very weak sources where the content of the analyte is very low. Another problem for spectrometric analysis arises when the peaks of various analytes neatly overlap and they cannot be easily unfolded. A typical example for such problems is in the analysis of γ-ray spectra from uranium or plutonium where special detectors and highly specialized software for the deconvolution of peaks must be used, or the analysis of α-spectra

from plutonium samples where one needs very special sample preparation techniques together with a very high resolution detector and highly sophisticated software.

Another problem that often hinders γ-spectrometric measurements is the presence of background counts which contaminate the measured data and can mask the desired peaks, or at least make calculated results statistically less significant.

Considering the pitfalls of nuclear spectrometry listed above, one may ask the question: why are its methods still in common use? The most important arguments are :

- the clear resolution in terms of element (Z) and isotopic mass (A)
- the relatively low cost of spectrometers
- the low detection limits
- the ability to resolve even very complex analyte mixtures
- the ease of sample preparation

An example from α-spectrometry will explain the detection limit situation. α-spectra are measured with high efficiency ($\leq$40%) and free of background counts; thus, every single measured count can in theory be assigned to an emitting nuclide. Even with a small peak of say 20 counts, one can calculate the peak energy and make an unambiguous assignment of the peak-counts to a nuclide. Taking ^{232}Th ($T_{1/2}$=1.45*10^{10}y) as the analyte, one will measure 59 counts in one hour from 10 μg of material. If the analyte is ^{226}Ra ($T_{1/2}$=1600y), then one will get 53 counts per hour from 1 pg and from 10 fg of ^{210}Po ($T_{1/2}$=22y) one measures 39 counts per hour.

In summary, α-spectrometry provides:

- extremely low detection limits
- extremely high sensitivity
- extremely specific (clear Z and A) analysis
- extremely low background

provided that the analyte has a suitable half-life. Very similar arguments apply for the usefulness of γ-ray spectrometry.

Development of nuclear spectrometry

The advent of drifted semi-conductor (germanium or silicon) detectors in the early sixties opened a completely new era of quantitative nuclear radiation spectrometry. Former scintillation detectors having reasonable spectrometric properties such as NaI(Tl) were rapidly replaced by Ge(Li) and Si(Li) detectors cooled with liquid nitrogen, and by diffused junction or surface barrier detectors for (α-)particle spectrometry in vacuum at room temperature. The next major step in the development of detector technology came with refined techniques for hyper-pure germanium material production in the early 80s that allowed the making of un-drifted HPGe detectors for penetrating radiation. At about the same time a novel technique of ion implantation allowed fabrication of particle detectors without the disturbing dead layer on the surface, leading to an improvement in resolution, and to cleanable (washable) detector surface.

Multichannel-analyzers (MCAs) used for NaI(Tl) scintillation spectrometry typically measured spectra with 256 and up to 1024 (=1k) channels, where peaks are sufficiently well resolved for quantitative analysis. High resolution Ge(Li) and HPGe spectrometry, however, requires at least 4096 channels spectrum length and modern spectrometers provide 8k or even longer spectra. The technology of MCA production advanced rapidly in the 50s after the development of semiconductors. Whereas the first MCAs were bulky and very heavy because they were operating with vacuum tubes and relay counters, the invention of transistors immediately transformed the MCA into a nice little stand-alone unit that could even be carried by just one experimentalist. With the advent of personal computers in 1981 and improved high-integration electronics, MCA technology migrated into the PC, and plug-in MCA cards were

developed that even contained the high voltage power supply for HPGe detectors, a spectrometry grade linear amplifier, a gain stabilizer and a very fast fixed dead-time ADC.

Quantitative spectrum analysis was made in the beginning via summing of counts and linear baseline subtraction for singlet peaks. Multiplets were analyzed through graphical peak separation and gravimetric integration. When mainframe computer centres were set up in universities, scientists began to develop a myriad of spectrum analysis software, only a few of which were actually very powerful and successful. Similar to MCA development, the advent of PCs changed the scene, and it brought another torrent of spectrometry software. However, the number of good software yielding reliable results did not increase significantly. Many programs assumed simplified Gaussian peak-shapes and linear baselines, whereas others excelled with over-complicated numerology without any relation to physics. Good software has been commercially available from various vendors since the 80s. It is interesting to note that several programs for the analysis of high-resolution gamma-ray spectra are available with a high level of sophistication but the selection of programs for the analysis of scintillator spectra is very meagre. This deficiency is particularly strange, because scintillation spectrometry is a frequently used method in many industrial and medical applications. All scintillator programs except one fit approximated shapes to the peaks and very few calculate the correct shape of the baseline. Similarly serious deficiencies are encountered in almost all programmes on the market for the analysis of high- or low-resolution alpha-particle spectra.

The following sections give an overview of current MCA and detector technology as well as of modern spectrum analysis principles.

MCA hardware

Since its appearance in 1981, the rapid development of PC bus technology has posed serious demands on producers of plug-in MCAs. Within only 29 years, the PC-bus appeared, followed by the ISA, EISA, MCA(=PS2), VESA, Local Bus, PCI, AGP for graphics, and finally PCI Express in different variations of transfer speed, to name just the most popular PC-bus designs. The function of plug-in MCAs is critically dependent on the bus structure, and thus for every new bus design, a new generation of plug-in MCAs had to be developed. Because of the relatively low production numbers of MCAs, the continuous redevelopment of cards was no longer profitable for producers and as a consequence, plug-in MCAs have disappeared from the market. Modern systems are rather external MCAs that link into the PC via USB or Ethernet or radiofrequency (RF) interfaces. Emulation software to handle spectrum measurement as well as data display and storage is almost exclusively based on one or several of the WINDOWS® operating systems. Three basic types of MCA presently dominate the market:
1- Stand-alone (or plug-on for scintillation detectors) units that contain all electronic functions but have no display to show instructions and measured data.
2- NIM MCA with or without built-in ADC hardware that are integrated into existing high quality NIM electronics.
3- Network MCA that are attached to the ADC in a NIM system.
Other designs are also available, still including plug-in cards with on-board ADC and/or multi-parameter capability.

As an example, a modern network MCA is shown in Figure 1 which is available for connection to almost every commercial ADC on the market. The unit of cigarette-pack size is powered via POE (Power-Over-Ethernet) and it is a true network MCA. Thus, the spectrometric measurement can be operated over any local or wide area network and an unlimited number of such MCAs can be operated from one computer.

The most important achievement of the "no nuclear electronics inside the computer" strategy is almost complete independence from future bus development. If any new interface definitions

arise, only the firmware or maybe the interface may have to be exchanged, but the MCA itself is retained.

Interesting properties dependent on new technology are often incorporated in current MCA design. Most units save a lot on power, thus allowing efficient operation from batteries or other weak power sources such as Power-Over-Ethernet or USB. This results in smaller and lighter MCAs with less cables to trip over, and which are portable and independent of mains power.

Figure 1:
Example of an Ethernet-based 64k MCA that can be directly connected to any commercial ADC

Spectrum length in modern MCA normally exceeds 4k channels and data are counted on 32 bits per channel. Some MCA emulator software and firmware are very user-friendly and allow intuitive handling of the MCA. Most functions are well described in an integrated on-line manual or in the bubble help. Unfortunately not all modern MCAs fulfil these desirable features. Finally, and most favourable for the buyer, the price of most modern MCAs is considerably lower than in the past.

In retrospect, it seems as if hardware development has turned full circle. The original MCA of the 60s was a complete stand-alone unit incorporating all necessary electronics, then came plug-in systems of various degrees of completeness and finally the modern MCA is stand-alone again. The computer is now only needed for MCA set-up, data display, storage and analysis.

The conversion in modern systems of analog current pulses from the detector into digital channel numbers for storage in a spectrum is achieved either by pre-amplification, amplification and digitalization of a well-shaped, large and slow analog signal with an ADC or by immediate digitalization of the small and fast preamplifier output with a flash ADC. The latter procedure is called digital spectrum analysis and it has overtaken the market in the last years. However, a few remarks have to be made on the status of converters: In many applications of nuclear spectrometry, the Wilkinson ADC is still the most suitable "workhorse" for stable and well-resolved conversion. The standard 100 MHz Wilkinson ADC developed in the 50's produces 8k-spectra with very reasonable dead-times for count-rates up to approx. 20 kHz. All ADC have incorporated dead-time control features; however, when very high precision of measured results is required, dead-time control and correction must be made by use of a precision pulse generator signal that is fed into the pre-amplifier. A proprietary method of "loss-free counting" was used for dead-time control of very hot samples or of samples that contain nuclides whose half-live is short with respect to the measuring time [1]. Despite the inherent problem of incorrect statistical significance of the spectral data, the loss-free-counting method is the only one yielding good results for fast-decaying nuclides in measurements of samples with count-rates up to about 100.000 counts per second. It is still of favourable use for certain applications in neutron activation analysis.

Some MCA units employ multiplexer-router logic in front of the ADC, thus routing pulses from several spectrometers through only one ADC. Routing techniques do actually substitute for several ADCs but at the same time they lead to dead-times in all channels that exceed the sum of individual dead-times from all spectrometers involved. Therefore it is highly recommended that multi-input systems that route all signals through one ADC should only be used in very low-count-rate applications such as e.g. in α-spectrometry.

There are undoubtedly several advantages of digital spectrometers over analog systems. The intercept of the energy calibration function is easily kept at zero thus enabling gain stabilization via only one peak. Due to modern processor technology, the count-rate throughput is as high as with a fast fixed dead-time (=successive approximation) ADC and certain firmware even allows correction for ballistic deficit and some crystal deficiencies. Despite these major advantages of digital systems, some caution is recently being adopted because of the somewhat worse resolution of digital vs. analog spectrometers. It may seem that the same HPGe detector yields better resolution when operated with an analog system than with a digital spectrometer.

γ-Detectors

The most important recent improvement in HPGe technology is the production of increasingly bigger detectors that reach relative efficiencies of over 150% now (relative to a 3"x3" NaI(Tl) detector). To achieve one percent of relative efficiency, one needs to have approx. 25 grams of hyper-pure germanium; thus a 150% single crystal detector weighs over 3.7 kg! For technological reasons, such a big crystal is very impressive and it is quite difficult to mount safely in the cryostat. For physical reasons such big crystal allows the measurement of gamma-rays having energies well above 10 MeV with good full-energy peak efficiency. Unfortunately, not all of its spectrometric properties are so beautiful. Due to long distances between photon interaction points and collecting electrodes, the pulses in very large HPGe detectors suffer from ballistic deficit effects which leads to peak-shape disturbance and resolution degradation in the measured spectrum. Digital spectrometers can actually correct for some of these problems. However, when searching for the optimum compromise for relevant parameters such as resolution, detection limit, efficiency, sample throughput and detector price, it turns out that the recommended HPGe crystal size for high-resolution and low detection limit gamma-ray spectrometry is around 40% relative efficiency. Two 40% detectors definitely outperform one 80% system in the sum of the above criteria.

In addition to the increase of the usable crystal volume, HPGe technology has produced several interesting and useful developments. Portable Dewar vessels with holding times between two and four days allow real in-situ high-resolution gamma-ray measurements far away from any mains power provided that a battery-operated MCA or a DC-AC converter is available that runs from the cigarette lighter of a car and drives the power supply. L-shaped cryostats are very well suitable for a set-up where the detector is mounted inside a small lead castle sitting on a table top in such a way that very easy access for sample changing is guaranteed. Special detector configurations can now be built where several crystals sit together in one cryostat or in one assembly in order to measure complex coincidences or geometric dependences. As an alternative, stacked crystals can be used to measure X-rays and gamma-rays simultaneously with seemingly one detector. The most advanced systems employing very refined HPGe configurations are crystal-ball detectors [2] built for 4π efficiency and position sensitive measurement of gamma-rays from high-energy nuclear interactions.

Other novel detector types such as e.g. Cadmium-Zinc-Telluride have not been able to take over the market, except in some niche market applications such as nuclear medicine or forensic sciences.

In the last few years, the resolution of NaI(Tl) and other scintillation detectors has improved for technological reasons. Whereas the "standard" 3"x3" NaI(Tl) detector typically had resolution of 7.5% (FWHM at 662 keV) a few years ago, essentially all detectors purchased nowadays have resolution specified and measured below 7%. 2"x2" detectors have even been measured to have 6.1% resolution. Significant improvement of NaI(Tl) resolution over the current figures is not expected because of physical restraints.

BrilLanCe detector

A new type of scintillator material has recently been developed on the basis of $LaCl_3$ or $LaBr_3$ crystals that are doped with 10% or 5% of cerium, respectively. The proprietary material is called BrilLanCe and in particular the $LaBr_3(5\%Ce)$ material named BrilLanCe380 (B380) is now being energetically marketed. Rapid development of crystal growing techniques for the hygroscopic material has now lead to crystals of up to 4" in diameter and 4" thickness. Spectrometric properties of the material are remarkably good and a large number of potential applications can be envisaged.

The most striking advantage of a B380 detector is its energy resolution. Good NaI(Tl) detectors have 6.3% resolution whereas good B380 are measured to have 2.8% resolution, i.e. a FWHM of 18.5 keV for the 661.6 keV peak.
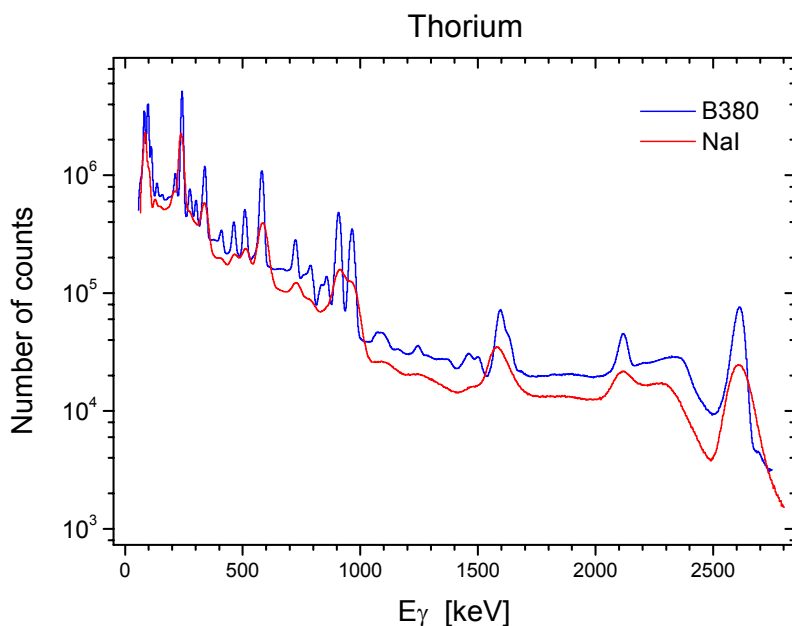
Figure 2:
Spectrum measured from a thorium source under completely identical conditions with a NaI(Tl) (lower spectrum) and a B380 (upper spectrum) detector

A comparison of measurements using identical 1.5"x1.5" NaI(Tl) and B380 detectors in exactly the same geometries and for the same live-time is shown in Figure 2.
Higher count-rates in the B380 spectrum as compared to NaI(Tl) indicate superior efficiency of the new material, especially at higher gamma-ray energies. Most striking, however, is the clear resolution of many peaks which are easily visible (and analysed) in the B380 spectrum but sometimes not even detectable with NaI(Tl).

Another striking example of B380 supremacy is shown in Figure 3, where part of a background spectrum measured under identical conditions and for identical times with NaI(Tl) and B380 is presented. A small contamination with ^{137}Cs is easily detected and analysed in the B380 spectrum whereas it is completely hidden in the NaI(Tl) spectrum. The lines in the B380 spectrum of Fig. 3 depict individual fitted peaks from [232]Th and [238]U progeny and the ^{137}Cs peak.

Another interesting feature of B380 detectors is its low sensitivity to temperature changes. The intrinsic light yield of B380 is very high and it hardly changes at all in the relevant temperature range between 20°C and 50°C. Thus, temperature changes that proved to be prohibitive for precise NaI(Tl) measurements do not disturb B380 spectra. Costly spectrum stabilisation is therefore not needed in most cases.

Scintillator light from B380 is not only very bright and stable, the signal is also very fast. Typical rise-times of B380 signals are on the order of a few hundred picoseconds, thus over 10 times faster than NaI(Tl). This feature allows very short coincidence resolving times and proper spectrometry at very high count-rates. The disadvantage of very fast pulses is a need for special
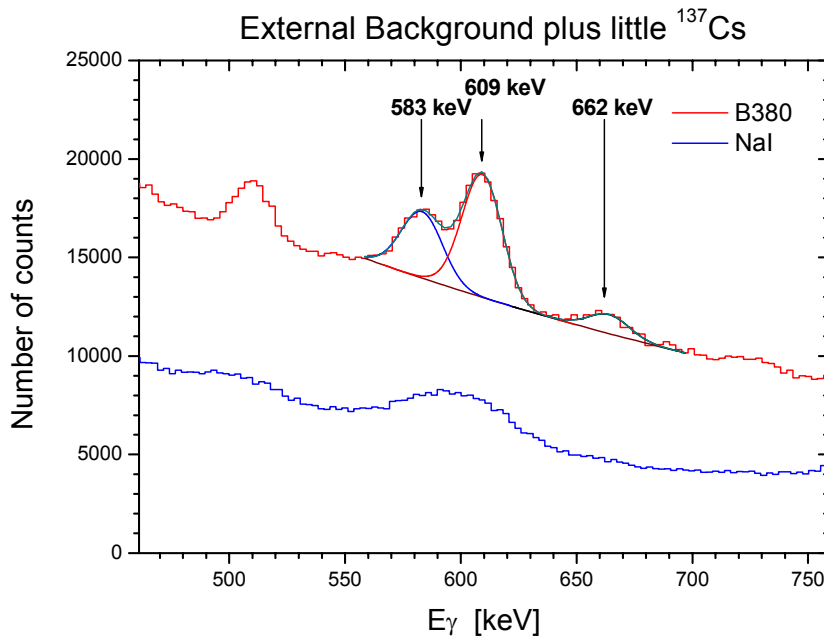
Figure 3:
Section of external background spectra measured with NaI(Tl) and B380 (top) detectors under identical conditions

input stages of spectrometry electronics for signal conditioning. This is, however, not a major drawback; it just increases the price of the system.

Due to its natural abundance of 0.0902% radioactive ^{138}La, the LaBr$_3$(Ce) material contains internal radioactivity amounting to approximately 139 cps in a 3"x3" detector. As ^{138}La decays by two modes, 66% β$^-$ and 34% EC, one finds gamma-ray peaks, Compton continua and Beta continua in the spectrum. Moreover, a small contamination of ^{227}Ac in the crystal material contributes to alpha peaks at higher apparent energies. This internal radioactivity, as shown in Figure 4, is a little annoying. However, as it is very constant (T$_{1/2}$ of ^{138}La is 1.05×10^{11} y), the background counts actually do not prevent or hinder quantitative measurements with the B380 detector.

When measuring the internal spectrum of a B380 detector in a very well shielded environment for a very long time, say 2 weeks or more, one may even apply pro-rata spectrum stripping in terms of energy (i.e. using precisely normalized spectra) in order to subtract the intrinsic contribution from a sample spectrum.

The peak around 1450 keV (see Fig. 4) is a doublet consisting of a 1436 keV gamma-ray and the coincidence peak of this energy with X-rays. The sharp rise above 750 keV is a 789 keV gamma-ray peak in coincidence with electrons from the 34% β$^-$ branch. Peaks above 1600 keV are caused by an internal contamination with ^{227}Ac which should decrease through improved crystal manufacturing technology in the future.

To conclude: New room-temperature scintillation crystals such as LaBr$_3$(Ce) have significantly better spectrometric properties than NaI(Tl). They provide huge advantages for many applications, such as better resolution, higher efficiency, higher light-yield, lower sensitivity to temperature changes, and better timing properties.
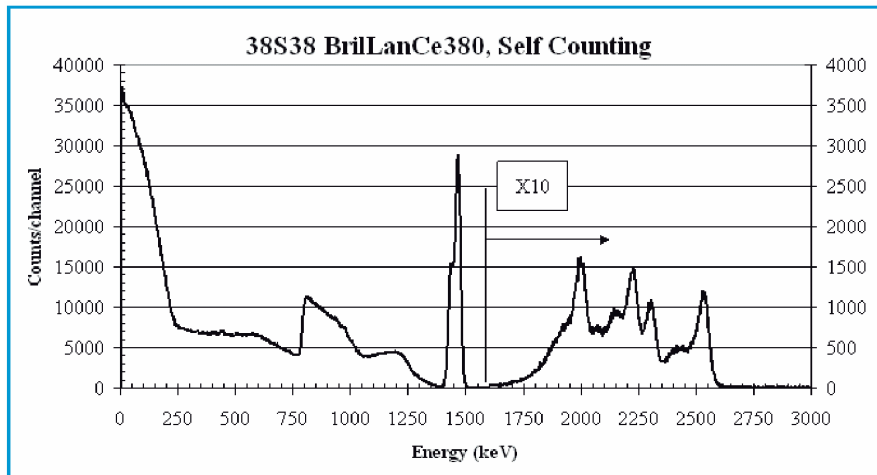
Figure 4:
Spectrum measured with a B380 detector from its internal radioactivity. The spectrum ordinate above 1600 keV is amplified by a factor of 10. (Courtesy Saint-Gobain Crystals)

At present, delivery times for B380 detectors are quite long and the price is quite high. This is expected to change. However, one should keep in mind that a B380 detector costs less than HPGe having the same efficiency for [60]Co. As an example, a 2"x2" B380 detector has the same peak efficiency for energies >100 keV as a 90% HPGe detector and it has the additional advantage that it needs no cooling.

Other non-proprietary materials such as, for example, Lu$_{1.8}$Y$_{0.2}$SiO$_5$(Ce) or LaBr$_3$(Ce) with <1% Ce are presently under development or are being tested and they may be expected to hit the market anytime soon.

α-Detectors

The last significant improvement in α-detector technology came when ion-implanted silicon detectors hit the market in the 80's. These detectors have better resolution than the older surface barrier detectors (SBD) and gentle decontamination of detector surfaces is now safely possible. Moreover these new detectors are hardly at all sensitive to corona discharge, thus allowing one to pump or vent the measuring chamber even when bias voltage is on. Because of the omission of a protective layer on the active detector surface the resolution of ion-implanted detectors is significantly better than that of SBD, now providing detector resolution <16 keV for partially depleted detectors with 450 mm^2 active surface and 0.3 mm thickness. Small detectors with resolution as low as 10 keV are now regularly available as stock items. The sensitive dependence of resolution on the distance between sample and detector has also been significantly decreased.

The technology of Gridded Ionisation Chambers (GIC) on the contrary has not made any recent significant advances at all. This neglect of GIC technology is a sad situation because GIC are the most suitable tool for many low-activity applications in α-spectrometry. Good GIC systems have resolution of better than 30 keV, depending on the quality of the preamplifier, and the efficiency of the chambers is always almost 50%. Moreover, the effective sample size is between 30 cm^2 and 300 cm^2, depending on the type of chamber used. Even when sample

preparation and/or the electronic setting of the preamplifier are poor, measurements with a GIC will yield valid results in short time and with low detection limits. As an example, the analysis of an α-spectrum measured with a 300 cm^2 GIC is shown in Figure 5. The mixed nuclide sample contained ^{239}Pu, ^{241}Am and ^{244}Cm of which the contents is perfectly quantified. Simple peak structures, such as the doublet of ^{244}Cm around channel 850 are even quantitatively correctly resolved by suitable software, yielding a fitted peak-area ratio of 3.19±0.56 as compared to 3.24 from literature data.
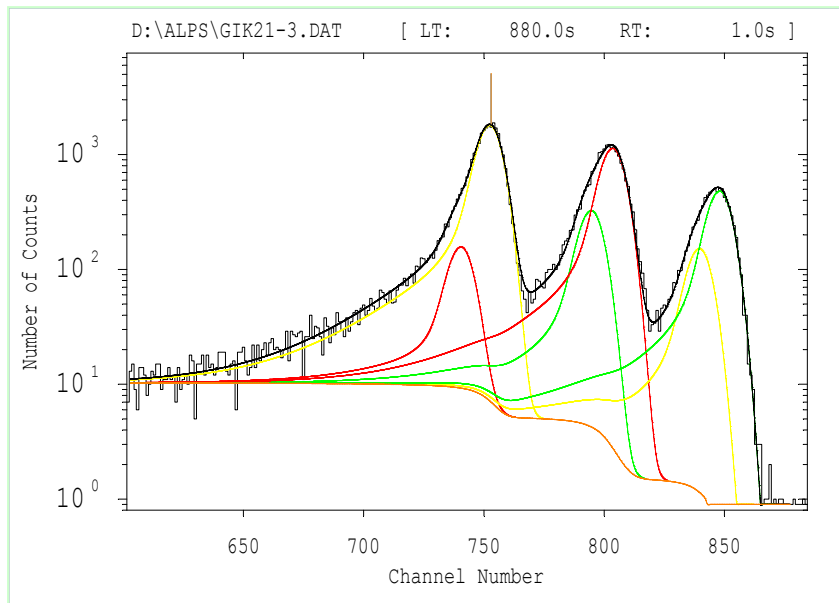
Figure 5:
Spectrum of ^{239}Pu, ^{241}Am and ^{244}Cm measured with a poor-resolution GIC detector and analysed with appropriate software

Although more complex multiplets, like the one from ^{241}Am around channel 800, cannot be resolved into their individual components, the total number of counts assigned to that nuclide is quantitatively correct. In Table I the count-rates analyzed from this spectrum are compared with the decay-corrected source data which are certified to within ±2%.

Table I: *Comparison of fitted data from GIC spectrum with reference data*

Nuclide	Peak counts	Relative intensity	Reference intensity
^{239}Pu	29321±1964	48.2±3.2%	49.14%
^{241}Am	21937±1909	36.1±3.1%	34.39%
^{244}Cm	9528±686	15.7±1.1%	16.46%

The sum-fit shown as a black line in Fig. 5, i.e. the sum of fitted peaks plus the underlying background distribution, matches the data very well. This figure demonstrates the measuring power and usefulness of GIC detectors very well.

Spectrum Analysis Software

After 1981, the development of software for the analysis of energy-dispersive spectra from nuclear decay has more or less followed the technical development of personal computers, and major emphasis has been on the user's request for simplified (intuitive) handling. All programs can analyse big singlet peaks correctly and with an acceptable level of accuracy, though it may seem that uncertainties of fitted parameters and of peak-areas are significantly underestimated in some programs.

A few general statements are:
  - All programs run under WINDOWS$^{®}$
  - All commercial programs have software protection units
  - All reasonable programs can read various spectrum formats
  - All reasonable programs can make reliable automatic spectrum analyses
  - All reasonable programs provide a software update service
  - All reasonable programs provide on-line help and hotlines
  - All reasonable programs provide spectrum oriented analyses but not peak-list
    oriented analysis.

Major inconsistencies are found in many programs where purely mathematical models are applied for the analysis of spectrometric properties and used to quantify peaks in the spectra (i.e. the numerology approach). Typical examples of inappropriate modelling are the linear or parabolic baseline under a peak, the purely Gaussian peak-shape, the square-root dependence of FWHM on energy, or the high-order polynomial description of the shape of an efficiency function for γ-rays. Correct spectrum analyses can only be made when physics-oriented descriptions of relevant spectrometric properties are found and applied in spectrum analysis. It is not meaningful to assume a straight line or another simple mathematical model for the shape of the baseline under a peak or multiplet. There is only one physical principle how the baseline is produced in the interaction of gamma-rays in the detector and there is only one shape of the averaged external background.

   As an example, we will take the baseline under a peak. It is clear that inclusion of counts that lie under the baseline of a peak will yield an excessively large peak-area and thus one has to separate the background counts prior to peak or multiplet analysis. This problem was first tackled by Pratt [3] and a review of commonly used nonlinear baseline shapes was presented by Helmer et al. [4] A graphical display of some of Helmer's examples is shown in Figure 6 where the peak-position is in channel zero.
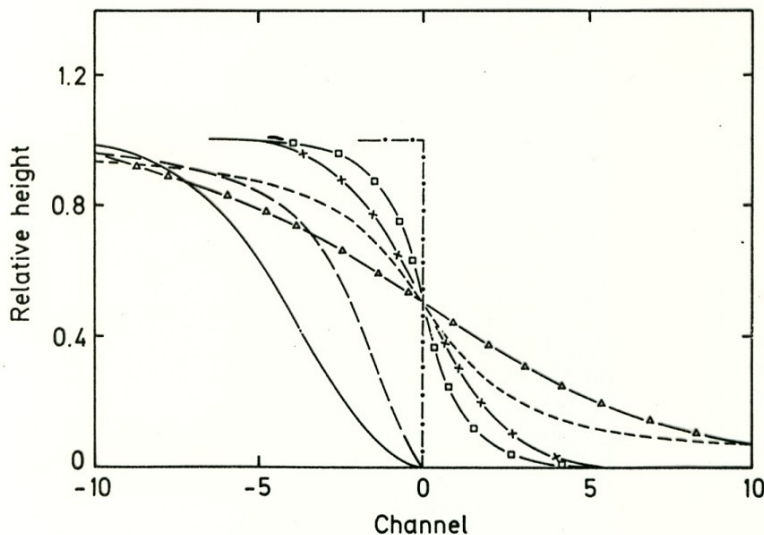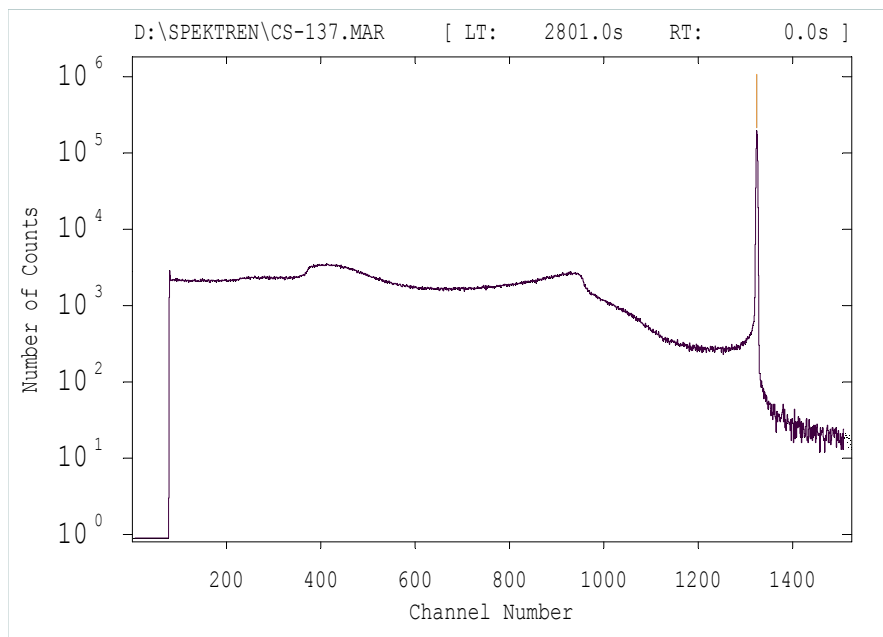

Figure 6:
Compilation of baseline-functions under a gamma-ray peak (after Helmer et al. [4])

Simple examples such as a straight line are omitted in the figure. In various papers it was later shown [5-7] how one can accurately calculate the correct shape of the baseline under a peak or multiplet exclusively from the numbers of counts in the channels in a region under consideration. The method works without any model assumptions or arbitrary peak-shape description. The procedure used is based solely on the physics-oriented finding that the background in the low-energy vicinity of a peak is constant, and it is smooth and continuous on the high-energy side, as shown in Figure 7. Moreover, there is a different height of the background before and after the peak, and the peak itself is almost a delta function. The method for baseline definition as

described in refs. 5-7 is based solely on these few simple and apparent facts. It allows one to calculate the baseline and subtract background counts from the spectral region to be analysed prior to the peak fitting procedure. There is no need to know the numbers, locations and shapes of peaks in the region where the background is to be determined. The difference between simple (mathematical) baseline functions and the correctly calculated one may amount to less than 0.3% of the peak-area of a large singlet peak. The importance and validity of the method proves itself when it comes to the deconvolution of multiplets, especially where a small peak sits on the shoulder of another big one. Only when the physically correct baseline is calculated and subtracted from the multiplet, will the fitted area of the small shoulder-peak match the correct value.

Another example of inappropriate numerology in peak analysis is the choice of purely mathematical descriptions of peak-shapes for the fitting process. Most programs adopt the physically supported assumption that the main contribution to a gamma-ray peak is a Gaussian distribution. A variety of model shapes are then assumed in order to describe the low-energy and sometimes also high-energy tailing of the peak. Such numerical approximations for peak-shapes without any reference to nature are not an acceptable solution because "nature does not throw dice" (cit. Albert Einstein: "Der Alte würfelt nicht").

Figure 7:
Spectrum with one singlet full-energy peak

The only acceptable procedure is to find out what the peak-shape actually looks like and then utilize that physics- or experience-oriented description in the spectrum analysis program. In this context, it should be noted that peak-shapes encountered for the old Ge(Li) detectors are different from shapes of modern HPGe detectors, and that the shapes found in very large HPGe detectors again may require different description.

A completely new strategy towards programming spectrum analysis has been introduced in recent years. The heart of the method lies in the fact that there are often various different ways in which a solution to a problem can be found. Some ways may be good for one situation but poor for another; some procedures are generally usable whereas others may be suitable for only very specific cases. This modern approach to handling the dilemma of "how to do it right?" is called FUZZY LOGIC where one:
- uses all available methods
- assigns weights to the different results, based on the applicability of the respective method

- discards inapplicable methods
- calculates the weighted average of valid results.

The averaged result of properly weighted individual solutions is almost always better, more stable and more significant than any single result. Typical applications in a spectrometry program for Fuzzy Logic solutions are, to give a few examples, the definition of regions in a spectrum that may contain peaks, the definition of the average height of the background before and after a peak or multiplet, the automatic generation of the resolution function, the assignment of nuclides to detected peaks, or the initial search for potential peaks and shoulder peaks.

A very difficult task is the definition of all peaks that can be fitted in a selected region of the spectrum. There are programs that deduce the list of possible peak candidates from the user-defined library. This method of library-oriented spectrum analysis, however, intrinsically has a serious deficiency. Nuclides that are missing in the library will never be detected and, often worse than that, missing peaks will lead to erroneous analysis of other peaks in the fitted region. The fully automatic analysis of a spectrum for determination of peak-positions and peak-areas basically requires a priori knowledge of all peaks that exist. As this premise is never fulfilled the program must analyse regions under the assumption that detected major peaks constitute all components. The residuum analysis of a fitted region may then reveal the presence of more peaks, or the analysis of statistical significances or peak proximity may render some of the fitted peaks as actually non-existent. A new fit of the same region but with a different set of potential peak candidates may yield an improved fit and finally, sometimes after three or four attempts to analyse a region, the statistically best possible solution will be found. Because of the very high computing power of modern PCs, such iterative region analysis is not a practical problem at all.
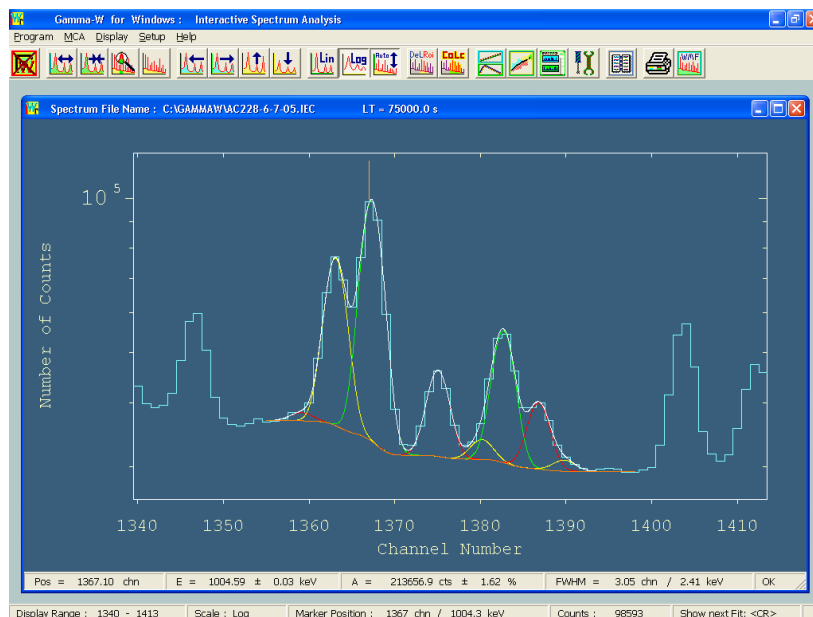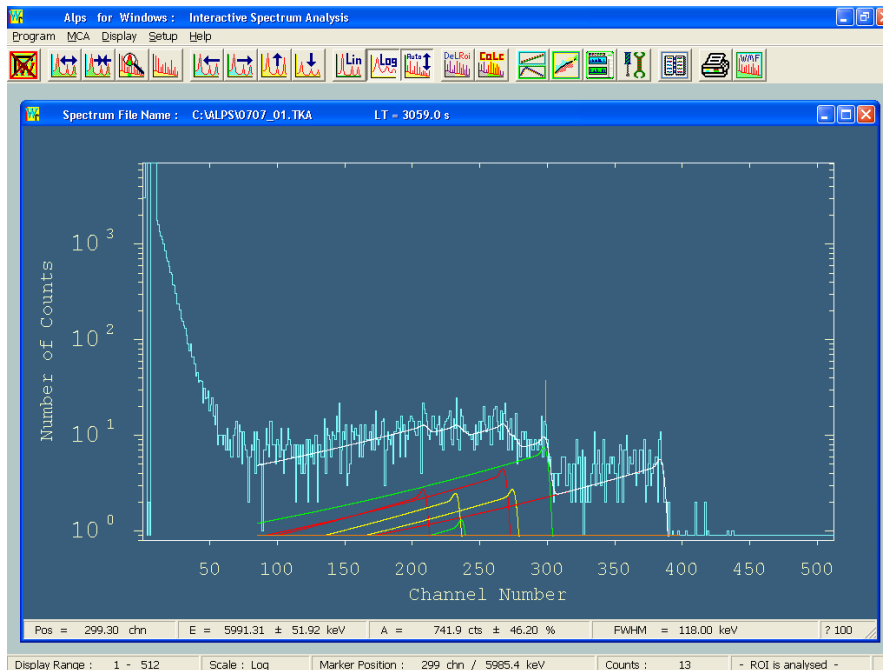

Figure 8:
Fully automatic analysis of a region in the γ-ray spectrum from ^{227}Ac

Examples of peak-fit results using commercially available programmes for spectrum analysis are displayed in Figures 8 to 10. A section from a HPGe spectrum taken from an ^{227}Ac source is shown in Figure 8 together with the course of the baseline and automatically fitted peaks. The sum-function of baseline plus peak contributions is also shown and it goes smoothly through the measured spectrum data shown as a histogram. As no peak-search algorithm will a priori find small peaks around channels 1359, 1380 and 1390, such complete automatic quantitative analysis of a region is only possible when consistent definition of peak FWHM with respect to neighbouring peaks or regions is controlled and when the region is subject to repeated analyses using various sets of possible peaks. Just to mention, the correct definition of uncertainties to

measured numbers of counts in the spectrum (considering robust statistics) is an essential aid for finding missing peaks without an over-definition of spectrum components.

In Figure 9 an unusual alpha-particle spectrum having very poor statistical precision is shown which was taken with an ion-implanted detector from a thin layer of "powder sample" consisting of grinded Pitchblende material. The average grain size of the powder was around 60-100 μm which is infinitely thick with respect to the range of alphas in material and therefore renders the material not suitable for high-resolution alpha spectrometry. Tails of peaks extend down to zero energy because alpha particles can be completely stopped within the thick sample. The maximum number of counts in one channel is around 20 counts, except in the low energy regime where beta particles and noise contribute to the spectrum.

Figure 9:
Analysis of an alpha spectrum taken from a sample of pitchblende powder containing ^{238}U *and progeny*

Using the physics-oriented peak-shape that is expected for such sample material, i.e. correctly considering the energy-loss of α-particles between emission and registration (which leads to a quite complicated mathematical description of the peak- and tailing-shape), one will find the fit presented in Fig. 9 where all alpha-emitting nuclides from the ^{238}U decay chain are identified and consistently quantified. Resolution into various component peaks for one nuclide as well as high-precision analyses yielding peak-areas with very small uncertainty are of course impossible in low-resolution spectra taken from powder material. The striking advantage of the new possibility to analyse spectra taken from powder or filter samples is the very ease of sample preparation. There is no more need for chemical dissolving of the specimen, element separation and deposition of a very thin sample. One can rather grind the sample material with a ball-mill or other suitable mechanical device and measure a thin layer of the powdered sample, or take a pressed filter as is, make the measurement and quantify analytes with reasonable accuracy.

In Figure 10 a region out of a ^{166m}Ho spectrum is shown which was measured with a BrilLanCe380 scintillation detector. The indicated region was automatically detected and analysed without user-intervention. Using the correct peak-shape description for this type of detector, proper FWHM description and calculation of the baseline according to the physical interaction of photons in that material the software can find and fit the region as indicated.

Peak-areas from all peaks having >5% intensity agree well within ±1σ uncertainty with literature values.
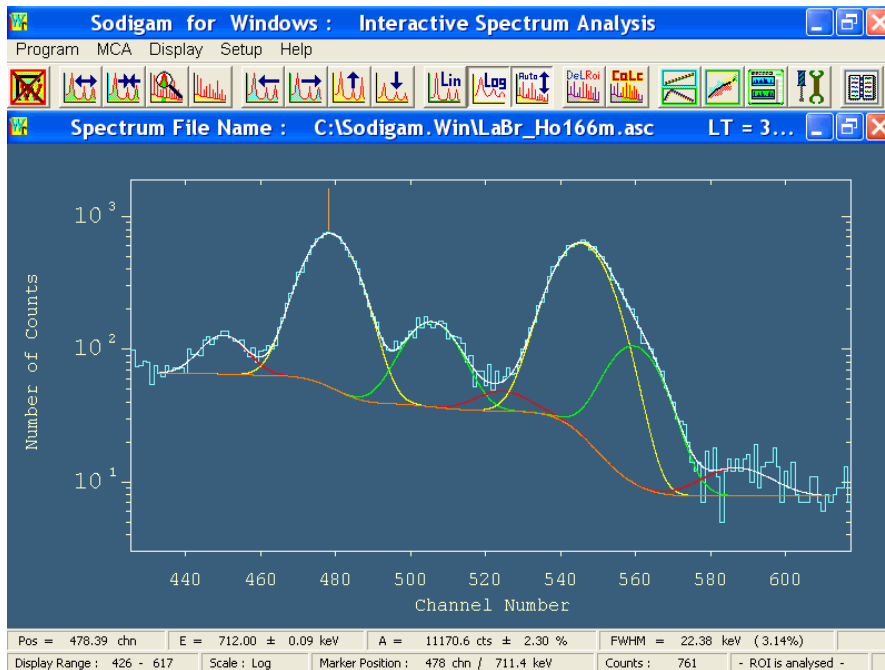
Figure 10:
Final result for a region around 750 keV of a ^{166m}Ho spectrum which was measured with a 2"x2" B380 detector and automatically analysed

Only through the consistent combination of methods and strategies described above, namely physics-oriented modelling, Fuzzy Logic strategies in solution finding and re-iterative analyses of regions can modern programs provide correct and fast analyses of spectra and yield reliable results for peak-positions and peak-areas of all peaks that can be analysed from the spectrum.
A very thorough and demanding investigation and inter-comparison of commercial and freeware PC-based γ-ray spectrum analysis software has been made some time ago by the International Atomic Energy Agency [8]. Only three out of the twelve tested programmes were listed, that "with respect to resolving power as defined in this test and quality of area determination .... yielded the least bad results".

Conclusions

The field of quantitative nuclear spectrometry is not dead. New developments of stand-alone MCA hardware make nuclear electronics completely independent from technical interface and bus progress in PCs. As a result, a new generation of MCAs is appearing on the market which are connected with the computer by wired interface or by radio-frequency. Network-based MCAs can be remotely operated and controlled from essentially every spot on earth and "distant experimenting" facilities are now feasible.

The improvement of resolution in traditional NaI(Tl) crystals and especially the development of new scintillator materials with significantly improved properties compared to NaI(Tl) has opened a completely new range of applications and possibilities. Many measurements can be made now with room-temperature scintillation detectors where the use of cooled HPGe detectors was once mandatory. Although the new B380 detectors will certainly take over significant parts of the HPGe-market, a large field of applications where cooled high-resolution HPGe detectors are indispensable remains.

Modern spectrum analysis programs use improved algorithms; in particular the modelling of peak-shapes, baselines and other spectrometric properties is based on physical correctness and not on numerical simplicity. The use of Fuzzy logic improves many tasks in automatic spectrum analysis and it allows much better definition of reference points and dynamic variables. Multiple

analyses of the same region using different reasonable sets of peak references are a good tool for successful result optimisation.

Finally, we wish to give an example of a system which shows that advances in modern nuclear technology can be fruitfully combined to make new applications: The combination of a modern scintillation detector with a plug-on MCA, a laptop PC and an up-to-date spectrum analysis program makes a complete portable gamma-ray spectrometer for in-situ measurement and analysis of samples. One can use such a system to measure and quantitatively analyse spectra from ores or soil for determination of its contents of uranium and thorium, or one can quantify nuclides in waste containers or other sources. Due to the very good resolution and high efficiency of BrilLanCe detectors one can now even use this room temperature scintillator in portable systems for measurements where very low detection limits and good resolution are essential.

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