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ABSTRACT

Different pulse processing systems have been tested at high counting rates for K-edge densitometry measurements with a continuous X-ray beam. The total input counting rates presented to the gamma detector have been varied between 10 kcps and 100 kcps. This paper describes the results of the measurements, in particular the influence of high counting rates on the spectral resolution, on the pile-up behaviour, on the throughput rate, and on the K-edge densitometry results.

Verarbeitung hoher Zählraten in der K-Kantenabsorptiometrie

ZUSAMMENFASSUNG

Verschiedene Impulsverarbeitungssysteme wurden bei hohen Zählraten auf ihre Eignung für die K-Kantenabsorptiometrie mit einem Röntgengenerator untersucht. Dabei wurden die Eingangszählraten am Röntgendetektor zwischen 10000 und 100000 Imp/s variiert. Der vorliegende Bericht beschreibt die Ergebnisse der Messungen, insbesondere werden die Auswirkungen hoher Zählraten auf das Energieauflösungsvermögen, das pile-up Verhalten und die Durchsatzrate der Meßsysteme sowie ihr Einfluß auf die Resultate der K-Kantenabsorptiometrie dargestellt.

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1. INTRODUCTION

In several applications of high-resolution gamma and X-ray spectrometry fairly high counting rates are available, but cannot be processed adequately by conventional pulse processing systems. At present the most common pulse processors employed in spectroscopy systems exhibit limited throughput capabilities, and they further suffer from the fact that the accumulated spectral data usually deteriorate with increasing counting rate due to a degradation in energy resolution and to pile-up effects. In many cases the detector input counting rates are therefore intentionally kept at moderate levels, say less than about 20 kcps, in order to maintain good quality of the spectra.

It is obvious that the capability to process higher counting rates is a desirable feature in many applications. The processing of high counting rates will reduce the counting time required for a given accuracy level and increase the statistical accuracy of a measurement within a given time. These aspects are of particular interest for in-line instrumentation purposes and for verification measurements by Euratom and IAEA as well, where the time available for the measurements usually is limited.

A spectrometry technique where high counting rates are easily available is L- or K-edge densitometry with a continuous photon beam from an X-ray generator. In our existing K-edge densitometers of this type /1,2/ we limit so far the counting rates to values around 20 kcps in order to maintain a linear instrument response versus the measured heavy element concentration. The reduction of the high beam intensity is achieved by means of a very tight beam collimation, which can be easily enlarged to yield higher total detector counting rates, if required.

In this paper we describe measurements where we increased the detector counting rates up to 100 kcps. For this study we employed different pulse processing systems, which included both conventional shaping amplifiers with time-invariant filters as well as an advanced processor with time-variant filters. With the various systems we evaluated characteristic parameters such as peak shape, pile-up rate and throughput rate. Finally, as one of the main objectives of this study, we investigated in more detail the impact of increasing counting rates on the K-edge densitometry measurements with the various pulse processing systems.

2. EQUIPMENT DESCRIPTION

2.1 Conventional Spectroscopy Systems

The measurements with conventional spectroscopy units employed a standard HPGe detector system. The detector with dimensions of $200 \text{ mm}^2 \times 10 \text{ mm}$ was coupled to a resistive feedback preamplifier with a maximum energy throughput of about 1.2 GeV/s.

The preamplifier signals were processed with conventional linear amplifiers providing semi-Gaussian (model Ortec 572) and/or triangular (model Tennelec 244) pulse shaping. Both amplifiers have built-in pile-up rejectors. For most of the measurements the shaping time constant in the amplifier was set to 1 µs. A few runs were also made using a shaping time constant of 0.5 µs.

The output signals from the shaping amplifiers were digitized in a Wilkinson-type ADC with a 400 MHz clock. The spectral data were converted into 4K channels and accumulated on a data acquisition and analysis system (ND 6600) with an add-one cycle time of 960 ns. Digital spectrum stabilization was employed by means of a dual-point digital stabilizer. The Np-La line at 13.9 keV and the 59.5 keV line from a ²⁴¹Am source served as reference peaks for zero and gain stabilization.

2.2 Fast Pulse Processing System

A fast pulse processing system, the Harwell Pulse Processing System NM 8000, has been temporarily obtained on loan from the IAEA, which previously had received the system for evaluation in the framework of the U.K. Support Programme. The additional field tests at KfK were carried out under an agreement of the Support Programme of the Federal Republic of Germany to the IAEA.

The Harwell Pulse Processing System received for the present measurements with K-edge densitometry represented a complete, autonomous spectroscopy system with carefully matched components. It included a $120 \text{ mm}^2 \times 10 \text{ mm}$ true planar HPGe detector coupled to a specially designed preamplifier using opto-type reset. Pulse processing in the advanced Harwell Processor is achieved by the use of time-variant filters /3/. These, together with comprehensive pile-up rejection, enable the processor to handle extremely high counting rates. The system is still operating up to input counting rates of 10^6 cps. The processing times are switchable between 0.5 µs and 10 µs, with the total dead time being approximately twice the processing time plus 1.5 μ s. We have selected a processing time of 1 μ s.

The analog-to-digital converter of the system provided conversion into 4K channels at a fixed conversion time of 4 μ s. Gain and zero stabilization control signals, which are fed to the pulse processor, are derived from digital selectors in the ADC. The zero control is set internally, while the gain stabilizer channel can be selected by the user. We have set the gain stabilizer on the 59.5 keV line from a ²⁴¹Am source.

The digitized data are stored in a derandomizer buffer for read out into the multichannel analyzer memory. The pulse processing system was equipped with a modified version of the CICERO-MCA from Silena. In order to enable the evaluation of the accumulated spectral data with our existing software routines running on the ND 6600 system, we established a special interface for data transfer to this system.

2.3 <u>K-Edge Densitometer</u>

The K-edge densitometry measurements were performed at a laboratory test facility. This laboratory set-up is of the same type as the K-edge densitometer, which is presently in use for safeguards verification measurements at the European Institute for Transuranium Elements, Karlsruhe /1/. The X-ray source consists of a voltage and current adjustable, highly stable 160 kV dc high voltage generator and a compact X-ray tube with thick tungsten anode, which provides an intense X-ray bremsstrahlung continuum. The necessary beam collimation was achieved by means of a 10 cm long, 0.2 cm diameter tungsten collimator located between X-ray tube and sample position. A second tungsten collimator, 1 cm long and 0.5 cm in diameter, was placed directly in front of the detector at a distance of about 15 cm from the sample.

A set of uranium nitrate solutions was prepared for the measurements with concentrations ranging from 50g/l up to 430 g/l. They were measured in glass cells with a path length of 2 cm. The transmission measurements were carried out by means of a suitably tailored X-ray beam with a maximum energy of 145 keV. The beam intensity was adjusted by varying the tube current to give total counting rates in the range from about 10 kcps up to 100 kcps. For the highest uranium concentration (430 g U/l) a maximum counting rate of about 90 kcps was achieved for the given beam collimation and filtering, which was kept constant for all measurements.

3. SYSTEM PERFORMANCE AT HIGH INPUT COUNTING RATES

3.1 Energy Resolution and Peak Shape

The energy resolution of the various pulse processing systems was investigated using a strong, uncollimated ⁵⁷Co source. Input counting rates between 10 kcps and 100 kcps were obtained by a variation of the source-to-detector distance. The full width at half maximum (FWHM), the full width at tenth maximum (FWTM) and the full width at fiftieth maximum (FWFM) were determined for the 122 keV peak in the ⁵⁷Co spectrum. Table 1 shows the FWHM, FWTM and FWFM values, and the ratios FWTM/FWHM and FWFM/FWHM measured with the different systems at an input counting rate of 10 kcps. Ideally, the gamma peak shape should closely approximate to a Gaussian curve down to one fiftieth of the peak amplitude. Therefore, for comparison, the ratios FWTM/FWHM and FWFM/FWHM for a true Gaussian shape are also given in the Table.

The graphs in Figs. 1 and 2 show how the peak-shape parameters obtained from the different systems degrade as a function of increasing counting rate. The percentage degradation plotted in the Figures is normalized to the values measured at an input counting rate of 10 kcps as given in Table 1. As expected, some degradation of the peak-shape parameters with increasing input counting rate was observed with all pulse processing systems under investigation. The measured degradation was smallest for the FWHM values (2 to 4 %), and largest for the FWFM values. This fact indicates a departure from the true Gaussian curve with increasing counting rate through the development of peak tailing effects. However, the values for the ratios FWTM/FWHM and FWFM/FWHM measured at 100 kcps were practically in all cases still within the acceptable range of better than 1.87 and 2.55, respectively, as recently recommended /4/. Nevertheless, the tailing effects should be taken into account in peak fitting algorithms, especially when dealing with the deconvolution of peak multiplets from spectra taken at high input counting rates. Tailing effects also play an important role in L- or K-edge densitometry measurements with continuous beams as discussed below in Chapter 4.

Planar Detector/ Preamplifier	Amplifier/ Pulse Shaping	FWHM (eV)	FWTM (eV)	FWFM (eV)	<u>FWTM</u> FWHM	<u>FWFM</u> FWHM
200 mm ² x 10 mm Resistive Feedback	Ortec 572 (1 µs) Semi-Gaussian	584	1066	1403	1.825	2,402
200 mm ² x 10 mm Resistive Feedback	Tennelec 244 (1 µs) Semi-Gaussian	589	1079	1427	1.832	2.423
200 mm ² x 10 mm Resistive Feedback	Tennelec 244 (1 μs) Triangular	573	1051	1387	1.824	2.421
120 mm ² x 10 mm Opto Reset	Harwell 8000 (1 µs) Time-Variant Filter	617	1131	1501	1.833	2.433
		True Gau	True Gaussian (theoretical): 1.823			2.376

Table 1. Peak-shape parameters measured at an input counting rate of 10 kcps.

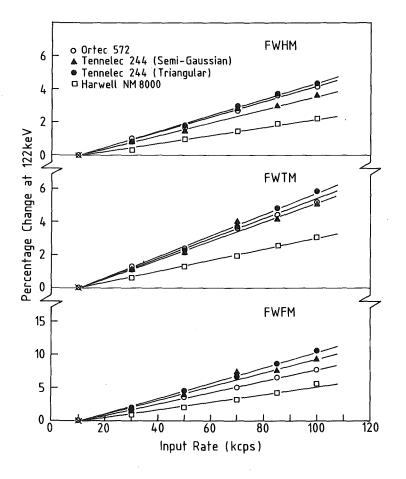
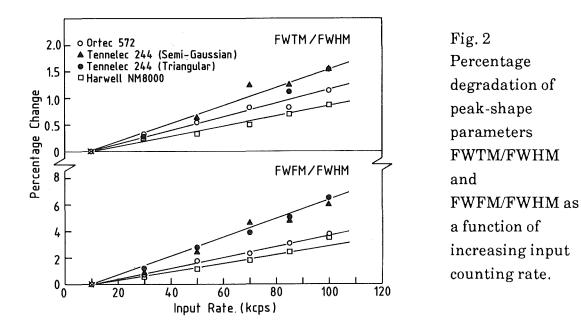


Fig. 1 Percentage degradation of peakshape parameters FWHM, FWTM and FWFM as a function of increasing input counting rate.

From Figs. 1 and 2 we note that the degradation of all peak-shape parameters was somewhat lower for the Harwell Pulse Processing System compared to the spectroscopy system equipped with conventional electronics. For an assessment of the obtained results one has to keep in mind, however, that the measurements were made with different detector/preamplifier systems, whose



specific properties may also influence the observed overall performance. For example, we noted that the detector used together with the conventional electronics showed a larger fraction of pulses with slow rise time than the detector coupled to the Harwell Processor. Therefore, for a true comparison of the performance of individual pulse processors a common detector/preamplifier system should be employed. This was not possible in the present measurements, because the Harwell Pulse Processor could only be operated in conjunction with its specially designed detector/preamplifier system.

3.2 System Throughput

The main parameter of interest in high-rate pulse processing is not primarily the maximum input counting rate that can be handled by the system, but the number of events really accumulated per unit time in the MCA memory. Fig. 3 displays the accumulated output counting rate versus input counting rate derived from the integrated MCA spectra and the real measurement time. In all cases the built-in pile-up rejectors were effective during the measurements. With the conventional electronics the input counting rate was determined by feeding the pulses from the count-ratemeter output of the respective linear amplifier to an external counter. In case of the Harwell Pulse Processing System the input counting rate was read from the digital ratemeter incorporated into the system.

The curves in Fig. 3 show that at the given shaping time constant of 1 μ s the accumulated output counting rate obtained with the conventional pulse processors reaches a maximum at about 25 kcps (Ortec 572) and 37 kcps

(Tennelec 244), whereas more than 60 kcps are actually accumulated with the Harwell Pulse Processing System. The slope of the throughput curve indicates that the maximum throughput for the latter system has not yet been reached at our highest input counting rate of 100 kcps. The dashed curve in Fig. 3 also shows that with a shorter shaping time constant of 0.5 μ s no significant improvement of the throughput behaviour of the Harwell System is obtained for input counting rates up to 100 kcps.

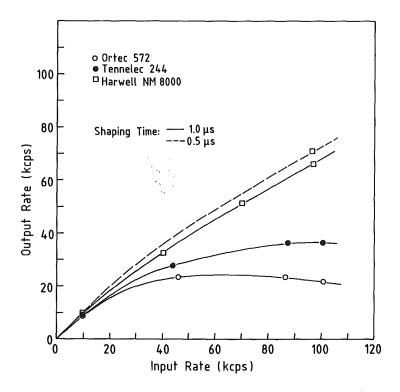


Fig. 3 System throughput as a function of input counting rate.

It must be stressed that the throughput rates given in Fig. 3 refer to the complete spectroscopy systems as described before. Therefore the measured throughput is also partly determined by the dead time introduced by the conversion time of the ADC and by the store cycle time of the MCA. There are additional parameters that influence to some extent the actual throughput such as the setting of thresholds, the selection of the pile-up inspection time in the pile-up rejection circuits, and the actual spectral distribution when Wilkinson-type ADC's with a variable, channel-dependent conversion time are used. The absolute figures for the throughput rates may therefore slightly vary depending on the actual experimental conditions and electronic modules used. Nevertheless, the curves displayed in Fig. 3 demonstrate that the Harwell Pulse Processing System is able to deliver significantly higher throughput rates than the conventional spectroscopy systems.

3.3 Pulse Pile-Up

Pulse pile-up or pulse summing is observed when the time interval between successive pulses is so short that the pulses will partly or totally overlap. This will result in a distorted pulse height analyzed by the ADC. The portion of pile-up events is a function of the pulse width, and it increases in a first order approximation linearly with increasing input counting rate. In order to avoid the distortion of the spectrum by pile-up pulses, most of the available amplifiers are equipped with pile-up rejection circuits that prevent the accumulation of detector pulses whenever two or more signals occur within a specified time interval. However, a complete elimination of pile-up events cannot be achieved due to the finite pulse-pair resolving time of the electronics, and also due to the non-zero threshold setting of the fast pulse discriminator. Therefore, if successive pulses occur within a time interval shorter than the pulse-pair resolving time of the pileup rejector, the sum of their pulse amplitudes is recorded in the spectrum. Also pile-up events with pulse amplitudes below the discriminator threshold are not recognized by the pile-up rejector.

The pulse-summing effect is demonstrated in Fig. 4 showing a typical K-edge spectrum taken at high input counting rate. Since the X-ray tube is operated at a high voltage of 145 kV, no primary X-rays with energies higher than 145 keV should be observed in the spectrum. All events recorded in the spectrum beyond this energy have therefore to be attributed to pulse pile-up. The pile-up spectrum was calculated from a simple model assuming summing of the channel contents below 145 keV (see Section 4.2). Fig. 4 shows that the calculated summing spectrum fits fairly well the structures observed in the spectrum above 175 keV. It can also be seen that the pile-up spectrum extends down to the energy region of interest around the K-edge. This suggests that the contribution of pile-up events to the background must be taken into account when measurements are performed at high counting rates.

The percentage fraction of pile-up summing events in the total stored counts, as calculated from the actual spectral data obtained with the 3 different pulse processors, is plotted in Fig. 5 as a function of the input counting rate. At 100 kcps input counting rate the total pile-up contribution ranges from about 3 to 5%. Though, in principle, these figures may allow to judge the quality of the pile-up rejectors in the different systems, it should be kept in mind that they were obtained with particular settings for pile-up resolving times and discriminator thresholds.

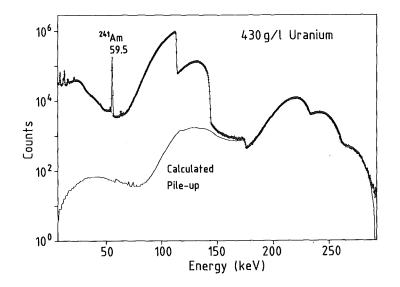


Fig. 4 Transmission spectrum and calculated pile-up background for an input counting rate of 100 kcps.

The minimum pulse-pair resolving time was about 500 ns for the pile-up rejection circuits in the Ortec amplifier and the Harwell Processor, and about 250 ns in the Tennelec amplifier. The shorter resolving time in the latter system explains its better pile-up rejection performance observed in Fig. 5. It should be mentioned that in the Harwell Processor another pile-up rejection circuit exists which gives an effective pile-up resolving time down to 100 ns or less. This option was not used for the present measurements.

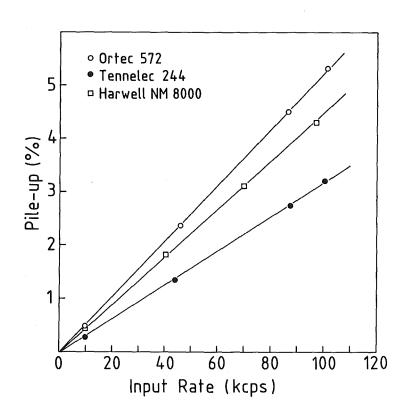


Fig. 5 Fraction of pileup events as a function of input counting rate.

3.4 K-Edge Densitometry

The major interest in the present investigations was devoted to the question, if and to what extent the K-edge densitometry result, i.e. the measured element concentration, is influenced by the input counting rate. To study this effect, we performed a series of K-edge densitometry measurements with the different pulse processors on the set of uranium solutions with concentrations between 50 and 430 g/l. With the 2 cm cells used, the effective areal uranium density of these samples covered the range between 0.1 and 0.86 gU/cm². The input counting rates were again varied between 10 and 100 kcps as for the other studies.

The spectral data were evaluated for uranium element concentration with our standard software using linear fitting techniques /5,6/. The program includes the subtraction of a step-like background below the broad X-ray peak prior to the determination of the transmission values in the K-edge region. At input counting rates up to about 20 to 30 kcps this background is predominantly a Compton background. Provisions for the treatment of individual background components such as pile-up are presently not included in the software for routine analysis.

The X-ray transmission is determined relative to a reference spectrum from a blank nitric acid solution. Separate reference spectra taken at an intermediate counting rate of about 50 kcps were recorded for each pulse processing system.

Some of the results are displayed in Figs. 6 and 7a. Fig. 6 shows the percentage change of the measured uranium concentration of the 430 g/l sample as a function of the input counting rate for the various pulse processing systems investigated. Fig. 7a displays the counting rate dependence at different uranium concentrations for a particular system. The results obtained with the Tennelec 244 amplifier are plotted in this figure. The other pulse processors exhibited a very similar behaviour.

The results in Figs. 6 and 7a clearly show that the measured element concentration decreases with increasing counting rate. This general trend has been consistently observed in all reported K-edge densitometry measurements using a continuous beam /5,7,8/. From Fig. 7a we note in particular that the counting rate effect increases with increasing concentration, which in fact means a non-linear instrument response. Under worst case conditions the non-linearity effect may lead to measurement errors in the order of a few percent, which is untolerably high compared to the accuracy of 0.2 % or better, which we obtain in routine densitometry assays at controlled moderate counting rates of about 20 kcps /9/. It - 11 -

has been experimentally verified that at this counting rate the non-linearity of the K-edge densitometer response is in fact negligibly small.

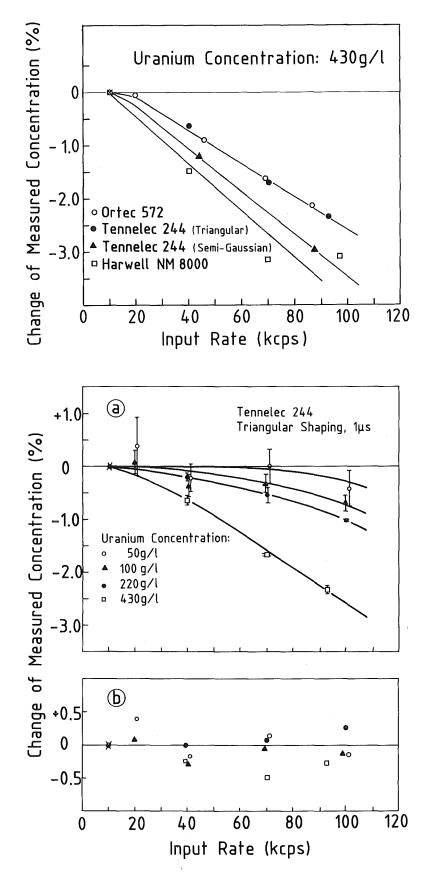


Fig. 6 Effect of input counting rate on element concentration measured with different pulse processors.

Fig. 7

a)Effect of input
counting rate on
K-edge densitometry results
for different
heavy element
concentrations.
b)Same measurements as in a)
after correction
according to
equation 1.

If measurements over an extended range of areal sample densities at higher counting rates are desirable, the measured results need to be corrected for the observed non-linear response. In principle this could be achieved by means of an empirical correction employing data such as presented in Figs. 6 and 7a. This has been done for the data given in Fig. 7a using the following empirical approach:

$$\rho_{\rm corr} = \rho_{\rm meas} \cdot (1 + 6 \cdot 10^{-15} \cdot \rho_{\rm meas} \cdot n^2), \tag{1}$$

where the corrected element density ρ_{corr} is derived from the density value ρ_{meas} obtained from our standard evaluation algorithm, and from the observed input counting rate n. The application of the correction to the data displayed in Fig. 7a is shown in Fig. 7b. A comparison of Figs. 7a and 7b demonstrates that the corrected assay results are less dependent on the input counting rate and on the element concentration in the sample, and that the relative error can be kept within the limit of ± 0.5 % for the particular measurement system and for the range of parameter variations investigated. More systematic investigations and more precise measurements may result in a better correction formula than the simple form presented in Eq. 1.

However, it should be pointed out that this pragmatic approach remains somewhat unsatisfactory as long as the true reasons for the non-linear instrument response are not well understood and the influence of various instrument parameters is unknown. Therefore we tried to identify possible sources of the observed non-linearity. In the following chapter we discuss some further investigations and thoughts on this subject.

4. BACKGROUND MODELLING

4.1 Problem of Background

The available experimental data suggest that the non-linear instrument response observed at high counting rates is related to the problem of correct background assessment. L- or K-edge densitometry is based on a transmission measurement, i.e. the value of interest is the number of X-rays with energy E penetrating the sample without changing their energy as compared to the number of primary X-rays of the same energy entering the sample. Any event falling into the spectral region of interest around the absorption edge, whose original energy has been altered, has to be considered as background. This background must be removed from the spectrum in order to arrive at correct transmission values. The background correction becomes particularly important for measurements on samples with a large areal density of the heavy element of interest, where the Xray beam above the absorption edge is strongly attenuated, thus leading to reduced signal-to-background ratios.

When dealing with narrow, isolated structures in a spectrum, as for example with well separated gamma peaks, the problem of the background determination is comparably simple: the background portion below those structures is estimated from an interpolation of the background values on the lower and higher energy side of the structure using relatively simple models for the background contribution below the peak (linear background interpolation, smoothed step etc.). In most cases this approach works sufficiently well because the interpolation of the background is usually limited to a narrow energy range. In contrast, when dealing with very broad spectral structures like the broad X-ray continuum in K-edge densitometry, the assessment of the true background becomes more problematic. Here the regions on both sides of the X-ray bump, in which the spectral data represent the actual background, are much larger separated (~ 80 to 100 keV) and simple interpolation methods with a global treatment of the background will probably not work for all experimental conditions. In order to arrive at an adequate description of the real background, it appears necessary to analyze carefully the various components contributing to the overall background continuum within the energy region of interest.

In the following sections we describe simple models used for estimating various components of the background.

4.2 Pulse Summing

In Section 3.3 we have discussed the effect of pulse summing. The summing spectrum N_{sum}(E) is calculated from

$$N_{sum}(E_1 + E_2) = a \cdot N(E_1) \cdot N(E_2),$$
 (2)

where $N(E_i)$ denotes the channel content corresponding to the energy E_i in the measured spectrum below the cut-off energy at 145 keV. The proportionality constant a is determined from a normalization of the calculated summing spectrum to the measured spectrum above 175 keV. As shown in Fig. 4 this simple model describes the summing spectrum fairly well. It also demonstrates that the summing spectrum contributes to the total background within the energy region of interest below the K-edge jump.

Further refinements to the algorithm may include summing effects of three or more pulses, and the iterative evaluation of the pile-up spectrum. However, these time-consuming computing procedures did not significantly improve the approximation of the summing spectrum, at least not for input counting rates up to 100 kcps as investigated in the present paper.

4.3 Compton Background

In gamma spectra, especially when taken with thick radioactive samples, one generally observes that the background at the low energy side of a gamma peak is significantly higher than on the high energy side. To give an example, Fig. 8 shows a portion of a gamma spectrum obtained from a thick ⁵⁷Co source at counting rates of 10 kcps and 100 kcps. The appearance of a step-like background below a gamma peak can be attributed to Compton scattering of the gamma rays within the sample material and within structural materials surrounding the detector.

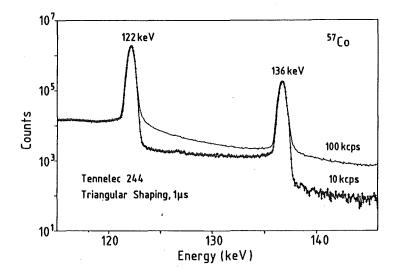


Fig. 8 Sectional display of ⁵⁷Co spectra taken at different input counting rates.

In our present densitometer we use a highly collimated beam geometry. Compton scattering in the sample (single scattering) is therefore only detected for very small scattering angles of 1° or less, at which the photon energy is practically not changed. Compton scattering of the transmitted X-ray beam in the detector itself leads to Compton events with energies below 50 keV, which do not interfere with the analysis. Further, since the collimated beam only hits the central part of the detector, scattering effects in structural materials around the detector are essentially limited to the entrance window of the detector cap and to the radiation shield. We therefore expect in our specific case that the contribution of Compton scattering to the background below the X-ray bump is relatively low. The spectral counts observed in the valley region at about 65 keV (see Fig. 4) are probably a good measure for the Compton background level in this region. Gunnink /10/ has proposed a method to evaluate the Compton scattering background from a measured spectrum. The model is based on the simple assumption that events accumulated in channel N of a gamma spectrum scatter a small, constant fraction of their intensity into all lower channels of the spectrum. The net counts and the scattering amplitude are determined by an iterative procedure observing the boundary conditions given by the measured background below and above the gamma peak. The resulting background has the shape of a smoothed step below the gamma peak.

The subtraction of the Compton background according to this method is implemented in our standard software for the evaluation of element concentrations from K-edge densitometry measurements.

4.4 Long-Range Tailing

When we compare in Fig. 8 the background shape of the two ⁵⁷Co gamma spectra accumulated with different input counting rates we clearly see that at high counting rates a long-range, exponentially decreasing tail on the highenergy side of the peaks is superimposed on the Compton background. Similarly we note from Fig. 4 that there exists an excess of spectral counts above the calculated pile-up curve in the energy region just above the endpoint energy of the X-ray beam, i.e. in the range from 145 keV to about 175 keV. This difference may also be explained by the existence of a long-range, high-energy tail originating from the X-ray bump below 145 keV.

In Section 4.2 we have only considered the case of completely overlapping pile-up pulses resulting in a pulse amplitude, which corresponds to the sum of the amplitudes of the two events involved. Partially overlapping pulses are normally rejected by the pile-up inspection circuit. However, the pile-up rejector operates only properly if it is assured that the pulse width does not exceed the pile-up inspection time. In many high-resolution gamma spectrometry systems one may occasionally observe very broad pulses at the amplifier output originating from bad charge collection in the gamma detector. Pulses with long tails can also originate from a bad feedback resistor in the preamplifier, from unsatisfactory pole-zero cancellation in the amplifier, and from an imperfect operation of the baseline restorer. We assume that the long-range tailing effect is presumably due to pile-up events with such long pulses.

In order to remove this portion of the background from the measured spectrum, we have extended Gunnink's model discussed in the previous section. It now includes the constant Compton background part on the low energy side and an exponential tailing component on the high energy side. The model parameters are obtained from the measured background shape at the boundaries of the X-ray bump. The pile-up summing spectrum has always been subtracted prior to the application of this modified background model.

4.5 Short-Range Tailing

In a further attempt to trace the problems associated with high-rate pulse processing in K-edge densitometry we calculated the ratio of two transmission spectra taken from the same sample at different input counting rates of 10 kcps and 100 kcps, respectively. We expect a constant value for this ratio throughout the whole spectrum as long as the overall spectral shape does not change with increasing counting rate. Fig. 9 shows the respective ratios for different uranium concentrations in the energy region around the K-edge. The ratios are calculated from the original spectra without any background subtraction. The dashed areas

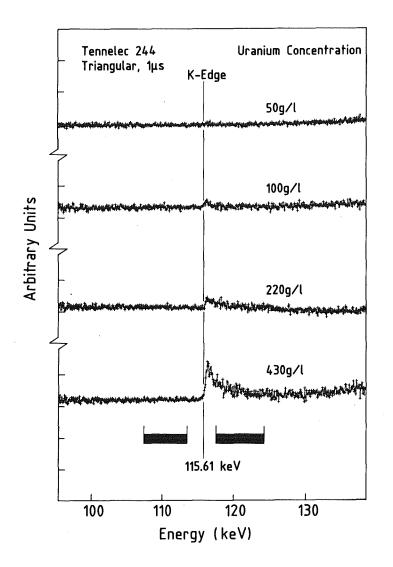


Fig. 9 Ratios of transmission spectra (high-rate to lowrate) for different element concentrations. at the bottom of the figure represent the energy regions used for the fits of the transmission values in our data evaluation /1,5/.

For the sample with lowest concentration the ratioed spectra yield practically a constant value within the displayed energy region around the K-edge. This picture changes with increasing element concentration. We observe an increasing deviation from constancy above the edge energy for the ratioed spectra. The shape of this deviation indicates that there is possibly a short-range tailing effect, which is not yet considered in our background model. It can be seen that this short-range tailing extends into the fitting region, from which we determine the transmission above the K-edge with our existing software.

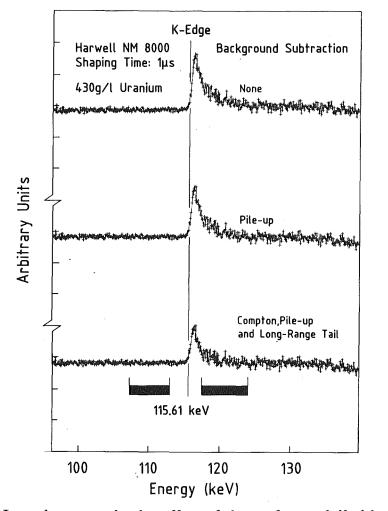


Fig. 10 Ratios of transmission spectra (high-rate to low-rate) after subtraction of different background contributions.

In order to study the effect of the so far modelled background components (Compton, pile-up, long-range tailing), we have calculated as before the ratios of spectra from low and high counting rates after various stages of background sub-traction. The results for the sample with highest element concentration are given in Fig. 10. In spite of small improvements with respect to the expected constant ratio of the spectra, the short-range tailing effect still remains after subtraction of

all background components considered. Therefore we have further modified Gunnink's background model to include now also a short range exponential tailing at the high-energy side of each net count considered. The exponential constant of this tailing component has been derived from the ratioed spectra shown in Fig. 10. The relative amplitude of the short-range tail was adjusted in such a way that the peaks just above the K-edge energy in Figs. 9 and 10 vanished.

4.6 Influence of Various Background Corrections on the Densitometry Results

It was of course interesting to see whether and to what extent the subtraction of the background below the K-edge jump influences the densitometry results. Fig. 11 shows a typical transmission spectrum along with the various background components discussed above. The measurement was performed at an input counting rate of 95 kcps using the sample with an uranium concentration of 430 g/l in a 2 cm cell. The different portions of the background were calculated according to the assumptions made in the previous sections.

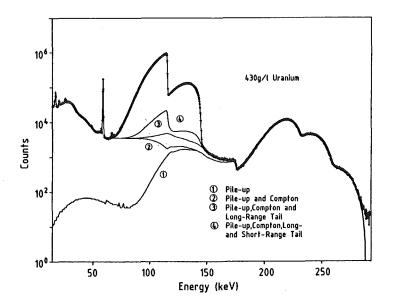


Fig. 11 Contribution of different background components to the transmission spectrum measured at 95 kcps.

Fig. 12 shows how the measured uranium concentration changes as a function of increasing input counting rate when different background components are included into the data evaluation. The trend and the magnitude of the observed effects were similar for all tested pulse processing systems. We note from the figure that, compared to the situation without background correction, the counting rate effect slightly diminishes when the Compton background is subtracted. The additional inclusion of the background due to pulse summing has only a

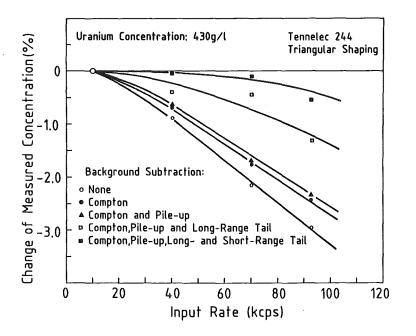


Fig. 12 Dependence of densitometry results on the background model used.

minor effect on the densitometry result. However, this may change if the input counting rate is further increased to values well above 100 kcps.

The largest improvement is obtained when the spectral data are corrected for the background arising from the long- and short-range tailing effect. The subtraction of all four types of background prior to the determination of the transmission ratio lowers the susceptibility of the assay results to variations of the counting rate and of the element concentration as can be seen from the upper curve in Fig. 12. The relative deviation of the evaluated element density is reduced to about 0.5 % after all corrections as compared to about 3 % without correction for the worst case condition of a sample with a large areal density of the heavy element (0.86 g/cm²) measured at an input counting rate of 95 kcps. The still remaining counting rate effect indicates that probably our background modelling does not yet adequately approximate the real background below the K-edge jump.

It must be stressed here that the evaluation of the various background components from the measured X-ray spectrum presents a difficult task. It requires a lot of computational effort, and the results in the present state of the evaluation method are still slightly susceptible to the selection of certain parameters, such as window settings. It should also be noted that the slope and the amplitude of the short-range tailing component are presently determined by measuring the same sample at two different input counting rates. This procedure is definitely not recommendable for routine measurements. One possible solution to this problem is to evaluate the tailing parameters at the beginning of a measurement campaign along with the basic calibration of the K-edge densitometer. However, this approach relies on the assumption that these parameters do not change for longer periods, and that they are unique functions of the input counting rate only. This has not been experimentally verified. Also the influence of the stability of crucial system parameters (e.g., threshold setting of the pile-up rejector) on the assay result and the impact of environmental conditions (e.g., line-voltage noise, microphonics) have not yet been investigated. Therefore, further efforts are required in order to extend the excellent accuracies of densitometric measurements presently achievable at low counting rates to high-rate applications.

5. SUMMARY AND CONCLUSION

The investigations presented in this paper show that high-rate pulse processing in high-resolution gamma spectrometry is possible today using commercially available electronic components. Throughput counting rates of 60 kcps or more are achievable with advanced systems such as the Harwell Processor at the expense of slightly reduced energy resolution and moderately distorted peak shape. However, all components of the spectrometry system, ranging from the detector to the MCA, must be carefully selected in order to meet the requirements of high-rate pulse processing. Due to the increasing interference of pulses in the analog electronics at high counting rates, special care must also be devoted to the proper adjustment of system parameters such as pole-zero cancellation and pile-up rejection, which are much less critical at low counting rates. In this context the use of preamplifiers with forced pulse reset offers the advantage to eliminate the need for pole-zero cancellation in the shaping amplifier. The adjustment for it was found to be extremely sensitive on the observed peak shape at high counting rates in the conventional systems using a resistive feedback preamplifier.

For the particular case of high-rate pulse processing in K-edge densitometry we found that significant assay errors may arise under certain measurement conditions, especially when samples with a wide range of areal element densities are encountered in the measurements. The observed counting rate effects most likely have to be attributed to the significantly higher background due to pulse pile-up as compared to low counting rate measurements. A wrong estimate of the background in the energy region around the K-edge may not only bias the densitometry results, but may also result in a deterioration and incorrect prediction of the measurement precision. Facing the importance of a correct background estimate in K-edge densitometry assays, a four-component background model was proposed that takes into account Compton scattering, pulse pile-up, short- and long-range tailing effects. The application of this model in the data evaluation procedure lowers the susceptibility of the densitometer response to counting rate variations to some extent. However, further refinements in the background modelling are needed.

In view of the fact that measurement accuracies of better than 0.2 % are presently obtained in routine K-edge densitometry assays at low counting rates, some further improvements in the hardware as well as in the software for data reduction are needed to arrive at a similar accuracy level with instruments operated at very high counting rates. Undoubtedly, the best solution would be given with further improved pulse processing systems, in which the various spectral distortions so far occurring at high counting rates could be eliminated at all through a proper design.

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