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FULLY AUTOMATED GAMMA SPECTROMETRY GAUGE OBSERVING POSSIBLE RADIOACTIVE CONTAMINATION OF MELTING-SHOP SAMLESS

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Introduction

In recent years, there has been a growing concern in the steel industry about the possible radioactive contamination of steel scrap ever since the first shutdown of the radioactive contaminated plant. We can easily foresee the increased exposure of steel market and steel mill as well as the risk of radioactive contamination.

These facts come along with the demands of some customers to supply steel "free of radioactivity". Of course, nobody can fulfil these strong demands, since natural radioactivity is always present. On the other hand, there are national and international limits for the release of material with respect to radioactivity. But so far these limits are still not levelled with all European countries. A detailed discussion is given by Wachtendonk (et al.) /1/ and Hoor /2/. Even for Germany release limits are still not completely determined. Mainly there are two types of limits, the specific activity with respect to the mass and with respect to the surface, which have to be checked.

By the theoretical reasoning, contamination of steel products can occur in the liquidifying process. Therefore only the specific activity to the mass [Bq/g] is of interest. The German Radiation Protection Regulation (Para.4, line 1) /3/ allows the handling of materials with a specific activity of less than 100 Bq/g for man-made radio nuclides and 500 Bq/g for natural radio nuclides by law. The German law for the transport of dangerous loads on streets /4/ and railways only allows 74 Bq/g. On the other hand there is a recommendation of the German Radiation Protection Commission /5/ which says that the sum of the specific activity of each nuclide divided by the limit of the specific activity of each nuclide must be smaller than one. Finally, now there is a recommendation for radiological protection criteria for the recycling of metals from the dismantling of nuclear installations "Radiation Protection 89" (1998) /6/, which refers to the Directive of the European Council No. 96/29/EURATOM from 13 May 1996 /7/ laying down the basic safety standards for the protection against the dangers arising from ionising radiation, which has to be transferred into national law. Although this recommendation does not contain the phrase of legal enforcement, it mentions a guideline for steel production in all.

Radioactivity and its Quantification

Radioactivity can be separated into alpha, beta and gamma activity. The different types of radioactivity and the corresponding measurement equipment are discussed by Wachtendonk (et al.) /1/. Alpha and beta activity can only be checked by such a large effort that observation during

production is impossible. Since many of these relevant nuclides also show gamma activity, the observation of gamma activity supplies sufficient information as a good approximation.

Gamma activity can easily be observed by scintillation counters. This technique also offers the possibility for spectrometry. Since gamma nuclides emit typical lines of gamma radiation, it is possible to determine the gamma emitting species and its concentration.

In the scintillation material gamma radiation is attenuated by effects of photoelectric absorption, by Compton scattering, and pair production. For all these processes electrons with a high kinetic energy are emitted which again lead to excitations of the adjacent atoms and then to recombination radiation which will be measured by photo multipliers.

When photoelectric absorption is dominant (e.g. for Ge- detectors), the emitted radiation can very well be determined. In figure 1 the spectrum of a Cs-137 source is displayed for different detectors. The Ge detector offers the best spectral resolution /8/. But, it must be constantly cooled by liquid nitrogen, which means large laboratory efforts. On the other hand

plastic scintillators show good results for the observation of large areas as a scintillation counter. A high spectral resolution is not possible as shown in figure 1c. NaI(Tl)-detectors offer a high quantum efficiency, which means that observation of the current production is possible with a sufficient possibility for spectrometry. An overview of the different gamma detector materials is given in table 1.

Table 1. Comparison of different detector materials

	Germanium	*Sodium Iodide*	*Plastic*	*Geiger-Müller*
Quantum efficiency	+	++	+	-
Time needed for observation	++	++	+	-
Spectral resolution	++	+	-	- -
Laboratory effort	o	++	++	+
Large area observation	O	+	++	o

The Salzgitter Concept

At Salzgitter AG there are several places where scrap delivery is checked, either wagonload or carload by large area plastic scintillation counters in form from 100% check, see figure 2. The observed data is automatically assigned to the corresponding wagon and afterwards stored in corresponding data bases for quality management reasons. In case of incident the Salzgitter AG fire brigade on guard will be informed and regulate the further measures. Alarm levels are about 10 % of the background radiation lowered by shielding effects

An installation for the observation of ships is foreseen. At the moment scrap from ships is reloaded to wagons, which are then checked by the rail observation system.

For ensuring the customers to fulfil their demands, a separate check must be performed with the produced material. Since no 100% guarantee can be given by observing the scrap delivery, at Salzgitter AG melting shop samples taken from the mould are checked for gamma activity. These samples are the samples for conventional release of the melt. In Salzgitter central chemical laboratory the samples are fed into the gamma spectrometry system after leaving the OE spectrometry line.

Salzgitter decided to work with NaI(Tl) detectors, in order to be able to calculate the specific activity of the samples, which is only possible by spectrometry measurement, since for each nuclide specific values of emissivity and attenuation coefficients have to be considered.

In figure 3 the complete and totally automated OE and gamma spectrometry line is displayed. The samples are coming from the preparation machine (right hand side) and are positioned on the OE spectrometry table by the robot. After determination of the chemical composition the samples are placed on the green coloured conveyor belt of the gamma spectrometry line. This conveyor belt works like a storage for samples and it is needed due to the different measurement times of the OE and the gamma spectrometer. Here, more then ten samples can be stored. Then the samples are placed into measurement position by a multi-linear robot (grey box). The gamma spectrometry gauge (red lead castle) consists of a sodium iodide detector and the necessary place for the sample positioned above, which are both shielded by 100 mm of lead from each side.

The amount of 100 mm of lead for shielding reasons is both, necessary and sufficient. On one hand, a reduction of lead thickness leads to an increase of the external background radiation. On the other hand, since natural activity of lead cannot be avoided, using more lead does not reduce the activity in the lead castle.

After 300s of measuring time the sample is removed. For quality management reasons and in order to allow for subsequent evaluation the relevant data is stored in a corresponding data base.

Calibration and Data Evaluation

Following the recommendation of the German Radiation Protection Commission /5/ and the European "Radiation Protection 89"/6/ the inequation

$$\sum_i {}^{A_i}\!/_{A_{\lim}} < 1, \quad i=1...n,$$

where A_i is the specific activity of each nuclide and $A_{\lim}$ is its upper limit, has to be fulfilled. Following /6/ the inequation may be simplified by conservative estimation $A_{\lim}$ >1Bq/g and by introducing the total activity

$$A_{tot} = \sum_i A_i < 0.1 \quad /g, \quad i=1...n.$$

Therefore, spectrometry is needed. Since not all nuclides can be evaluated, from a technical point of view the most relevant nuclides will be considered (Na-22, K-40, Co-57, Co-58, Co-60, Ba-133, Cs-137, Ir-192, Pb-212, Am-241). They are chosen to cover most of the spectrum between 55 keV and 1.5 MeV. In principle, some of these elements cannot be found in steel, therefore in case of incident these nuclides just give an indication of the typical energies occurring in the spectra. For each nuclide the detection limit can be calculated from the background spectrum. In figure 4d a typical background spectrum is displayed.
For determination of the specific activity the system has to be calibrated. The attenuation and geometry effect of NaI(Tl) must be considered (figure 4c) as well as the quantum efficiency (figure 4a). The factor representing the quantum efficiency varies a lot among the relevant nuclides. In figure 4b the typical spectrum for the different calibration nuclides (Co 60, Cs-137, Ba-133) is displayed. The calibration of the system can be checked e.g. by K-40 test specimen which are easy to prepare.

The gamma spectrometry system works with a two step calibration. The first step is the calculation of the detector efficiency curve (figure 4c), which is checked regularly for quality control. The second calibration step considers the energy drift of NaI(Tl) detectors. Therefore, before measuring the sample an energy calibration is performed by evaluating the photoelectric peak of a small Cs-137 source, which is positioned in the shutter of the shielding castle and can only be seen by the detector when the shutter is opened half the way.

For quality management reasons all relevant data starting with the sample number, the date, the measurement time, the specific activity for each nuclide, the total specific activity as well as the total count rate, and the corresponding detection limits are stored.

The alarm limits are set at 0.1 Bq/g, 1 Bq/g, 5 Bq/g, and 100 Bq/g. Furthermore, it is possible to calculate a dose rate for the situation that in a steel plant a man works in a distance of 1 m away from the bottom centre of a potentially contaminated 200t ladle. An alarm limit is set to the dose rate of 7,5 µSv/h with respect to the German law /3/.

Results from the gamma spectrometer at Salzgitter

At Salzgitter AG now both gamma spectrometry lines run for approx. two years. Figure 5 shows a typical cps- plot for the last four months. In total approx. 2cps are measured when the lead castle is empty or when no activity is in the sample. For measuring times of 5 min the statistical variation of the background gross counts is in the range of 4%, which corresponds to statistics for radioactive decay.
For every sample the measured specific activity was below the detection limit of 0.05 Bq/g.

Literature

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/8/ Knoll, Glenn F., Radiation detection and measurement, 2nd edition, John Wiley & sons, USA 1989.

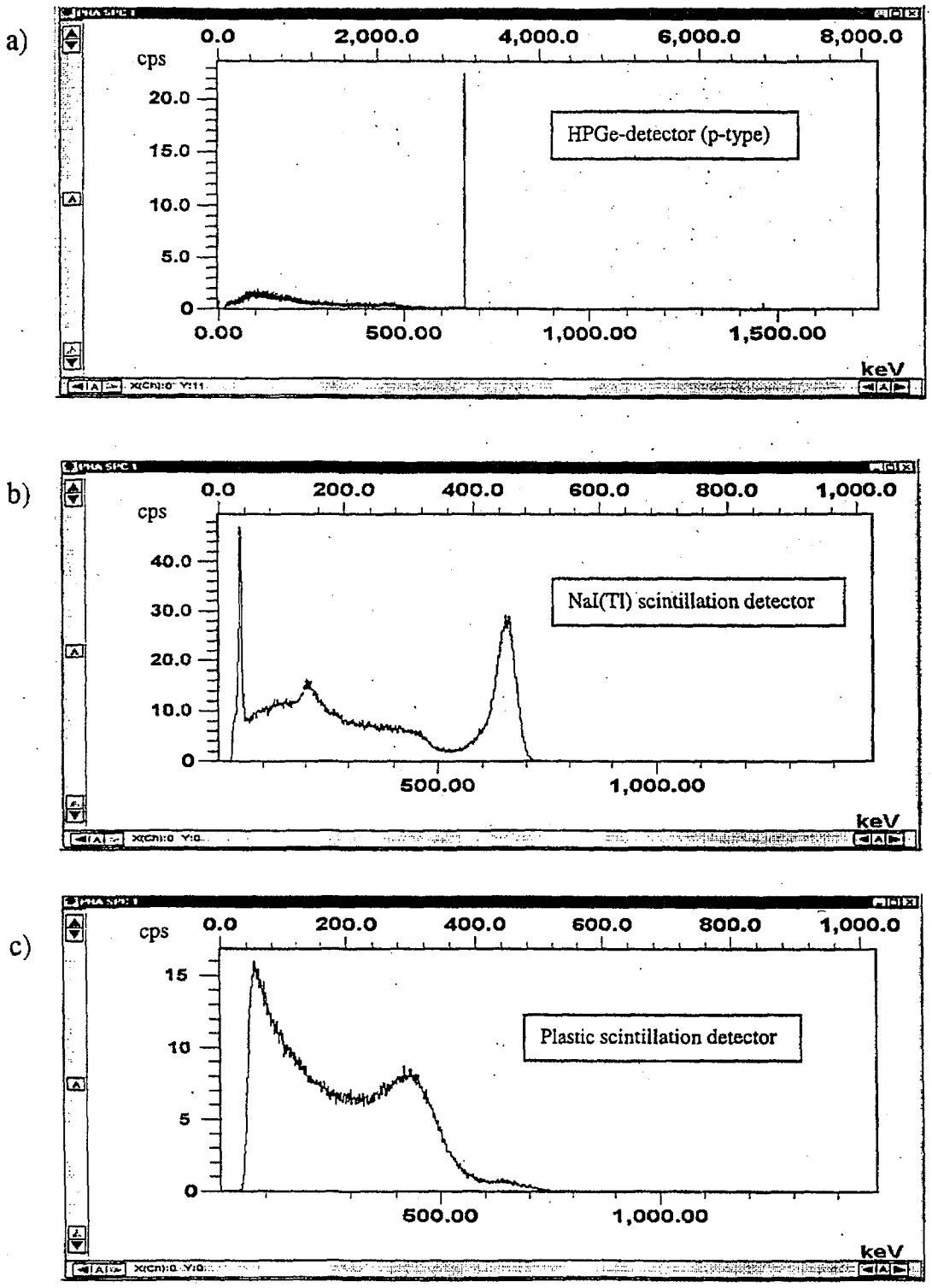

Figure 1. Comparision of pulse height spectra using a Cs-137 source
The comparison of these three different detectors shows the superior energy resolution of the Ge detector system. Due to the limited photo peak efficiency, a nuclide identification cannot be performed with a plastic detector. The NaI detetctor is used when the number of isotopes to be identified is limited.

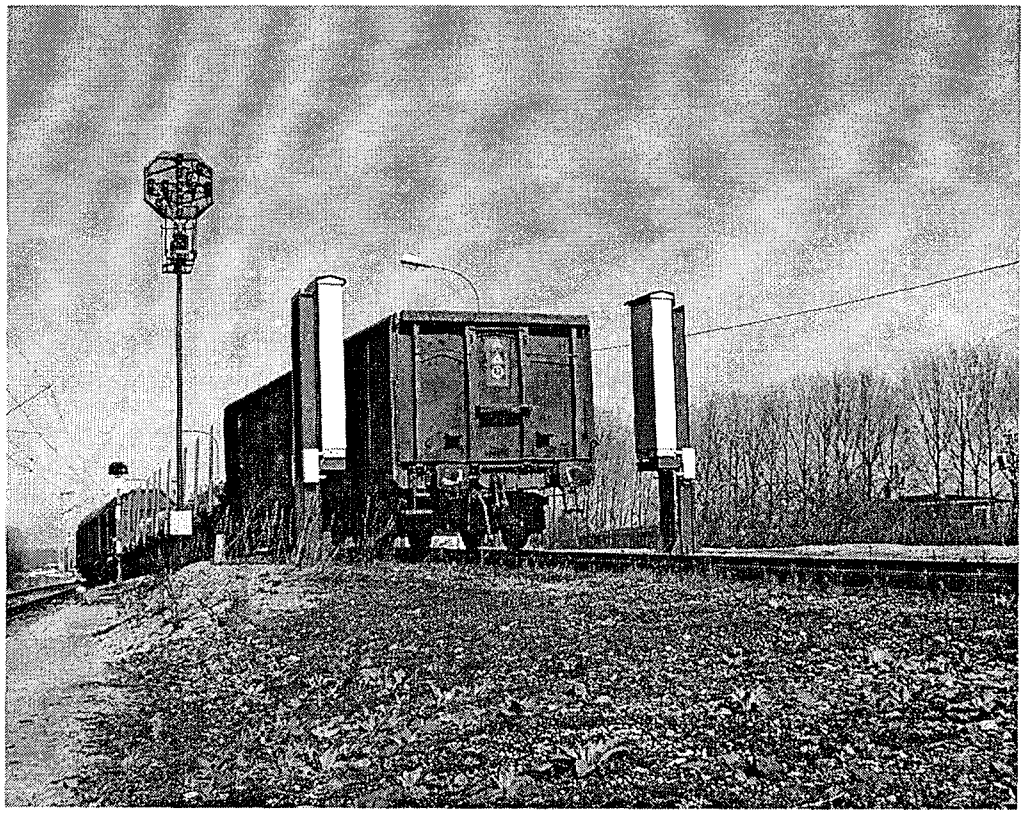

Figure 2: Wagon and carload check point

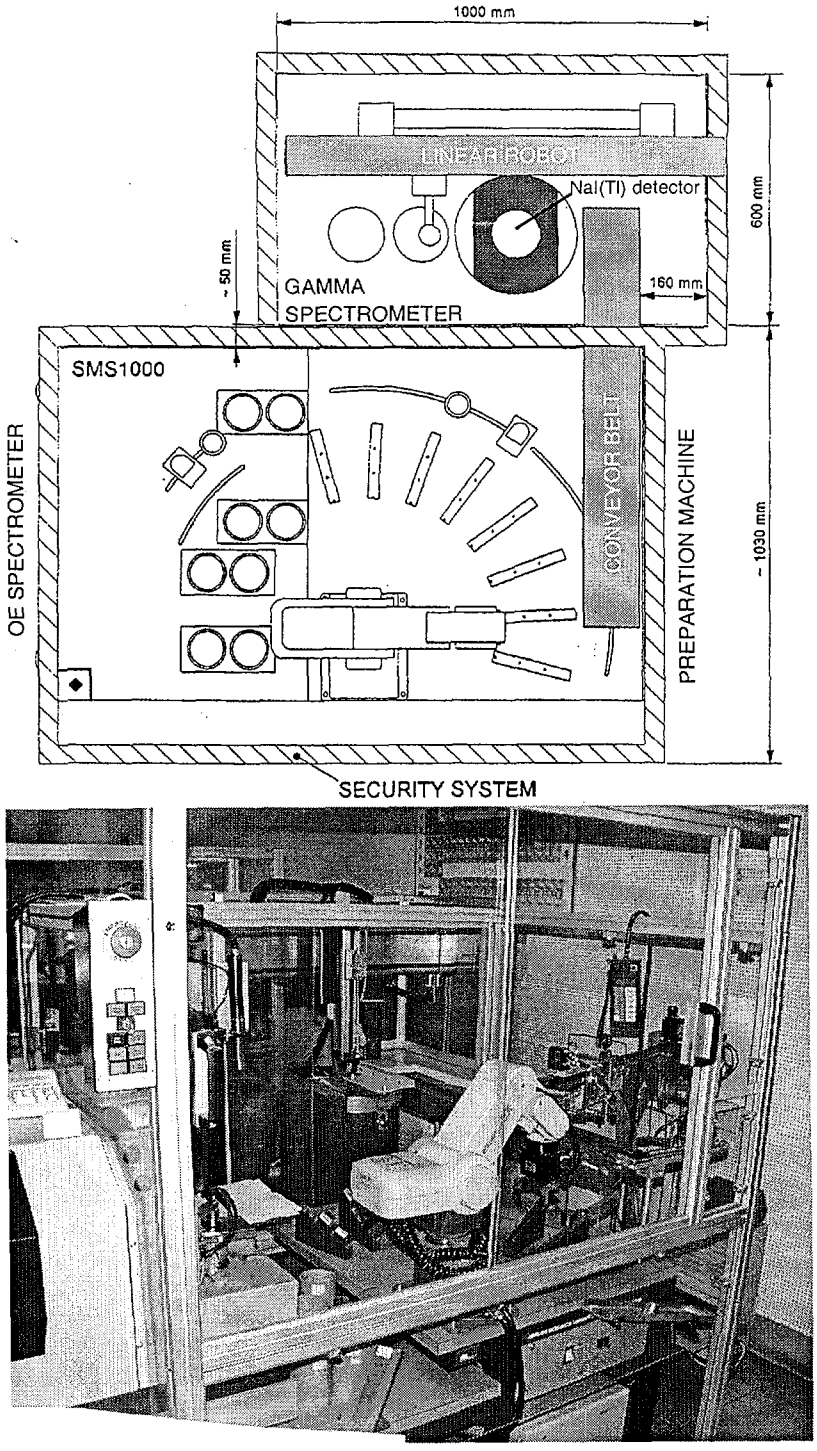

Figure 3: Sketch and photo of the automated gamma spectrometer integrated into the OE spectrometer line

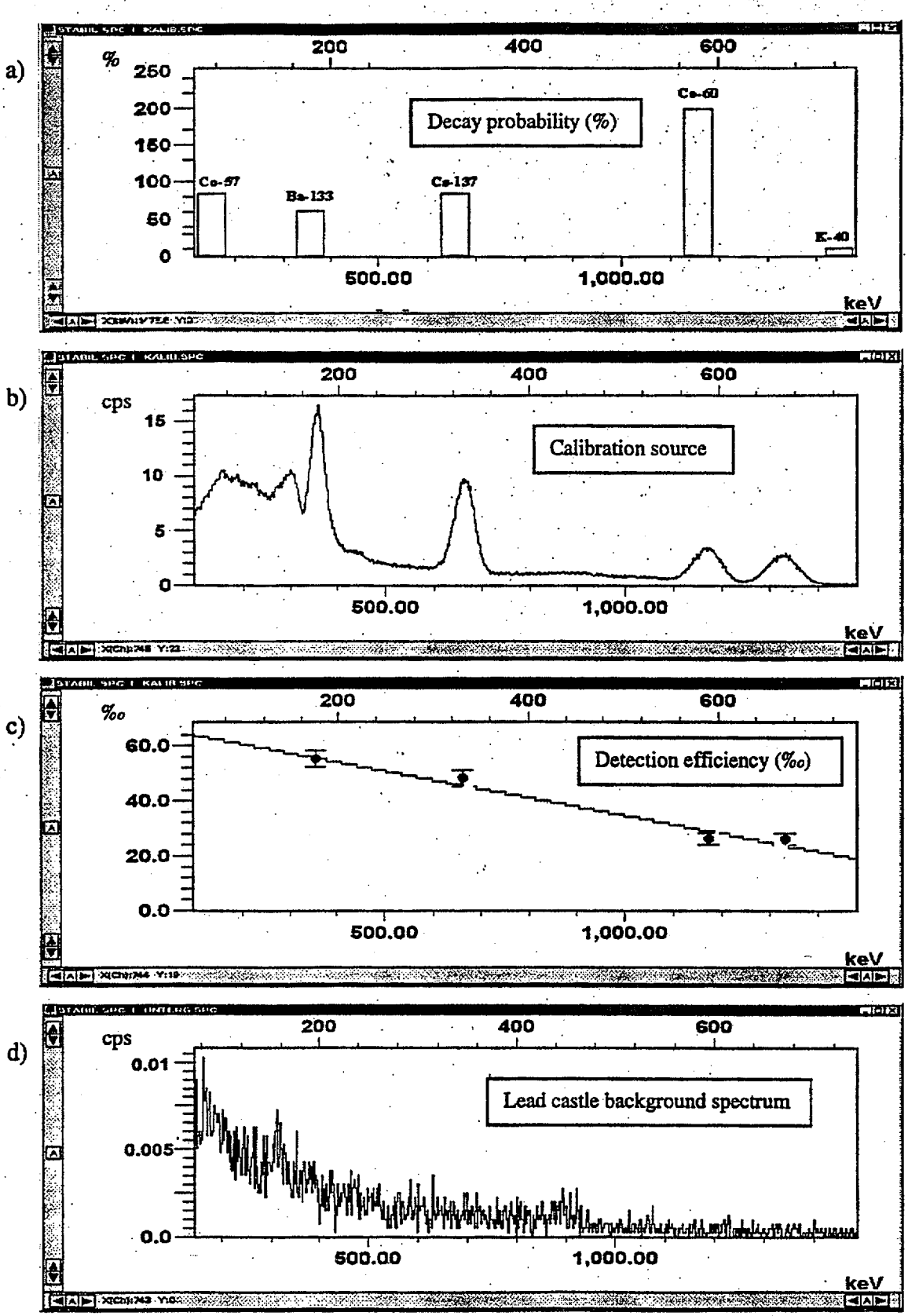

Figure 4. Activity calculation in NaI-spectra

The activity in Bq is calculated by a multiplication of three factors. The detector efficiency is multiplied by the net photo peak area and the decay probability. A relation of the absolute count rate for one nuclide cannot be extended to different nuclides as the decay probabilities differ by one magnitude. The total detection limit in Bq is given by such calculation for a background spectrum

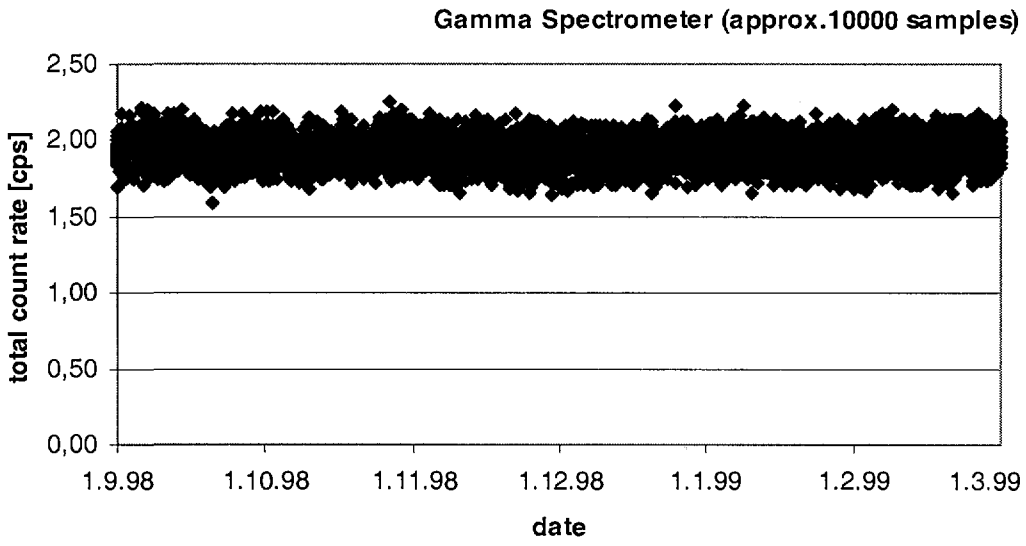

Figure 5. Total count rate for different samples (background count rate 2 cps)