Acta Physica Hungarica 59 (1—2), pp. 103—106 (1986)

DETERMINATION OF BETA-EMITTING IMPURITIES IN RADIOACTIVE CERTIFIED REFERENCE MATERIALS TO BE USED AT NUCLEAR FACILITIES

T. BARTA, S. LUEFF, Á. SZÖRÉNYI
National Office of Measures (OMH)
1531 Budapest, Hungary

The radioactive certified reference materials (RCRMs) have a very important role in the calibration of radiation protection instruments at nuclear power plants (e.g. surface contamination monitors, semiconductor spectrometers). In addition to the activity (or radioactivity concentration) as certified values, the radioactive impurity is also an important characteristic of RCRMs. In our office a suitable semiconductor beta-ray spectrometer has been developed for measurement of beta-emitting impurities. A short review will be given of the construction and the main characteristics of the beta-ray spectrometer. The computer aided evaluation of beta spectra with the method of "spectrum-splitting" will also be outlined. At last the results of simulation tests and the impurity determination of different radioactive sources will be discussed.

Introduction

One of the most important characteristics of radioactive certified reference materials (RCRMs) is the impurity which has so far been limited usually to the determination of gamma-emitting impurities only. To determine other kinds of impurities is a much more difficult task, nevertheless laboratories preparing RCRMs devote more and more energy and attention to the determination of beta- and alfa-emitting impurities, too. This paper discusses the OMH semiconductor beta-ray spectrometer as well as our results in the field of beta-emitting impurity determination.

Si(Li) semiconductor beta-ray spectrometer

The semiconductor beta-ray spectrometer consists of a measuring chamber, vacuum-system, temperature control and electronic signal processing system [1].

Block diagram of the spectrometer is shown in Fig. 1. One semiconductor detector or two facing each other can be placed in the stainless steel measuring chamber. The detector is cooled by cooling fluid stored in a tank above the chamber through a copper cooling rod of 20 mm diameter. Copper-constantan thermocouples are fitted on "U" shaped bridle connected to the cooling rod, and the temperature of the detector during cooling down and warming up is monitored by these thermocouples. A collimator can be placed between the detector and the source to be measured. The chamber has several openings (insertion and removal of the source etc.) that can easily be closed. A scintillation or semiconductor gamma-ray detector is inserted through the largest opening, and thus emitted X- and gamma-ray photons can also be measured simultaneously with beta-particles. Si(Li) semiconductor detectors of large depletion layer (2-5 mm) are used to the measurements.

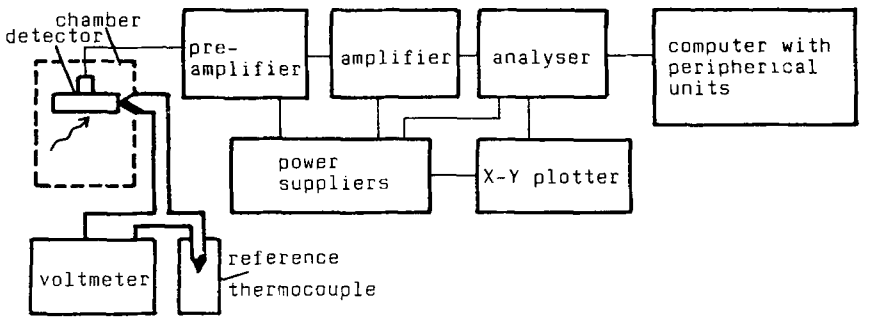

Fig. 1. Block diagram of the beta-spectrometer

Vacuum necessary to the measurements is produced by three sorption pumps and one ion-getter pump.

The temperature control comprises copper-constantan thermocouples and the thermal electromotive force of the measuring point is indicated by digital voltmeter. Thermocouple supplying the reference voltage is immersed in melting ice. Cooling is made by means of constant-temperature mixed fluids.

Canberra Model 2003B preamplifier and Model 2021 main amplifier is connected to the detector in the electronic signal processing system. Canberra Model 8100 4096-channel amplitude analyser applied to gamma-spectrometry, too, is used for signal analysis. Measured beta-spectra are processed further by DEC Model PDP 11/05 computer connected to the analyser.

The energy resolution of the beta-ray spectrometer and the counting efficiency stability was determined by special series of measurement. ^{137}Cs, ^{207}Bi, ^{204}Tl and ^{89}Sr sources were used to the tests. For a 5 mm depletion layer Si(Li) detector the energy resolution related to 624.2 keV conversion electrons of ^{137}Cs was 12.1 keV. The stability of the energy resolution was constant within the statistical uncertainty. For ^{204}Tl and ^{89}Sr sum of counts in the whole energy spectrum was measured several times within three weeks. Change in total count rate was less than $\pm$ 2 % for ^{204}Tl, and $\pm$ 1 % for ^{89}Sr.

Evaluation of beta-spectra

During the evaluation of beta-spectrum by means of Fermi-Kurie plot the measured beta-spectrum shall be decomposed according to degrading maximum beta-energies. If after the subtraction of Fermi-Kurie plot residual spectrum still occurs, this may be caused by the impurity. This impurity can be determined by a new Fermi-Kurie plot that is plotted to the residual spectrum. The new plot gives the maximum beta-energy of the beta radiation of impurity and then the relative intensity of components can also be determined based on the Fermi-Kurie plot "area".

The shape of beta-spectrum measured by the semiconductor detector deviates extremely from the shape outlined by the Fermi-theory - for reasons that cannot be given here in detail - therefore the evaluation is very elaborated involving a number of manipulations [2], [3], [4].

In OMH less complicated methods of impurity determination have been developed for the time being. Under strictly defined and constant measuring conditions we try to collect pure so-called reference spectra free from impurity and to store sources like that. Beta-spectrum obtained during the measurement of the source is compared to the reference spectrum by using different methods of calculation. Measured beta-spectra are normalized to equal measurement time and activity. Beta-spectra are described by the following experimental relation which is fitted by the least squares method

$$\ln(B) = a + bX + cX^2 + dX^3 \qquad (1)$$

where $X = \ln(E/E_1)$ and $E_1 \leq E \leq E_2$

E_1 and E_2 are the lower and upper limit of the energy range; B is the counts measured at energy E; a, b, c and d are constants.

After subtraction the reference spectrum or the spectrum described by the relation (1) from the beta-spectrum measured by the source occurrence of residual spectrum hints at impurity. A reference spectrum of approximately equal end-point energy is fitted to the residual spectrum and this will be subtracted, too. This procedure is continued until the spectrum disappears in accordance with the statistical fluctuation. Based on this composition determined as first approximation reference spectra corresponding to the basic and impurity nuclide is mixed until it complies with the initially tested spectrum within the statistical uncertainty of spectrum measurement.

The method of so-called "spectrum-splitting" evaluation outlined hereafter was tested with several kinds of nuclide. Beta-spectrum measured during the impurity testing and the reference spectrum is split up in 30-40 keV energy ranges and the total count rate in each energy range is calculated and then the ratio of total count rates relating to the successive ranges are calculated, too. These ratios practically "describe" the shape of the spectrum. The percentage of deviation of the above ratios relating to the measured spectrum and reference spectrum is then calculated. From the deviations obtained versus energy can be defined if it is an impurity of maximum beta-energy lower or higher than that of the basic nuclide, as well as the approximate maximum beta-energy of the impurity. The deviation is proportional to the impurity content. Based on this preliminary testing we can determine such much larger energy ranges whose total count rates can be used for more precise calculation of the impurity. This procedure was tested by simulation tests, too. Different kinds of beta-spectrum measured by beta-ray spectrometer were mixed in various proportions by computer and the obtained so-called mixed spectra were analysed. We have analysed e.g. ^{89}Sr mixed spectra containing 0.35-17.5 % ^{45}Ca impurity as well as 0.04-4.5 % ^{90}Y impurity. Fig. 2 shows curves of typical deviation

from the reference spectrum obtained in two tests. Fig. 2.a shows an impurity of maximum beta-energy being lower than that of the tested nuclide. Fig. 2.b shows impurity of maximum beta-energy higher than that of the tested nuclide. We have also evaluated limiting values for detecting the impurity [1] which are for certain nuclides as follows: (2.7 ± 0.3) % for ^{35}S impurity in ^{204}Tl, (2.1 ± 0.2) % for ^{45}Ca impurity in ^{89}Sr, (0.4 ± 0.05) % for ^{90}Y impurity in ^{89}Sr and (0.04 ± 0.008) % for ^{89}Sr impurity in ^{45}Ca.

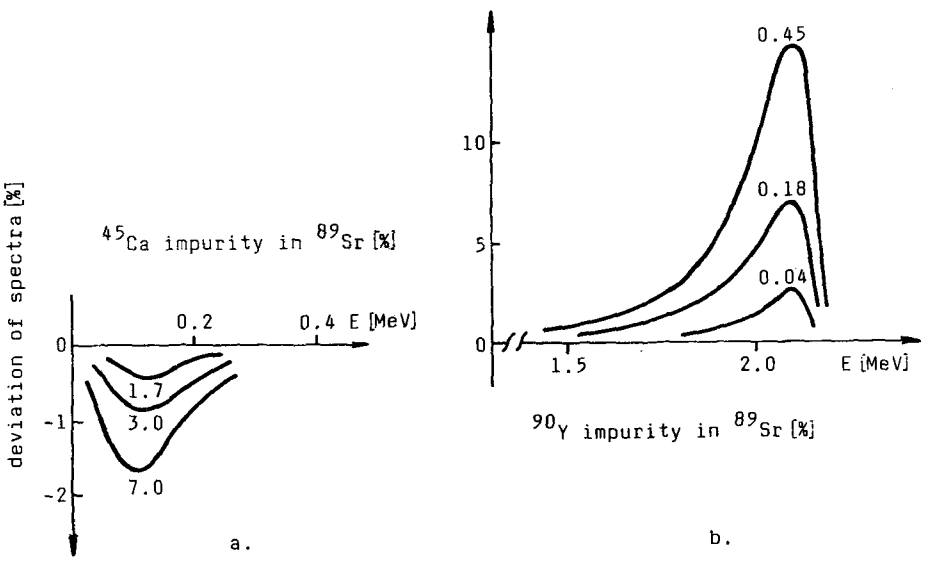

Fig. 2. Spectrum deviations depending on impurity content and energy

Tests were made with sources prepared by our Laboratory for Radiochemistry. The radionuclidic composition of these sources was unknown for us, e.g. a source that contained about 10 % ^{204}Tl impurity in ^{45}Ca. By means of beta-spectrometry (8 ± 2) % impurity was determined. By a more precise analysis have showed that ^{45}Ca master solution contained initially about 1 % ^{46}Sc impurity. Therefore we have dealt with the testing of solution separately and detected 1 % ^{46}Sc impurity by beta-spectrometry, too, in accordance with the previous result of gamma-spectrometry.

References

1. T. Barta and S. Lueff, in Proc. 2nd COMECON Symp. on RCRMs. ed. Á. Szörényi, Atomic Energy Comm., Budapest, 1984. p. 47 and p. 77 (in Russian).
2. G. Bertolini et al. Nucl. Instr. Meth., 27, 281, 1964.
3. P. Dryak et al., UVVVR report, No. 5., 1977.
4. V.R. Bom, Nucl. Instr. Meth., 207, 395, 1983.