

COLLIMATED IN-SITU GAMMA SPECTROMETRY: A NEW METHOD FOR FAST CLEARANCE MEASUREMENTS OF LARGE AREAS OR BUILDING STRUCTURES OF NUCLEAR FACILITIES UNDER DECOMMISSIONING ¹

Lothar Hummel; P. Guglhör; K.H. Lehmann
Technical Inspectorate of Bavaria (TÜV Bayern Sachsen, München)

INTRODUCTION AND MOTIVATION

The ideas in the late sixties which led to the use of bare spectrometric radiation detectors like NaI(Tl), Ge(Li) or nowadays high-purity germanium detectors (HPGe) for field measurements (in-situ gammaspectrometry) were to get easy and rapid informations about the radiological state of outdoor grounds after nuclear weapon tests or to estimate dose rates created by natural radioactive nuclides in the soil. In this cases, it was assumed, that there was no disturbance of the source-detector geometry for many hundred square meters around the detector. After the nuclear accident in Chernobyl in 1986 these advantages focussed strong scientific interest at the in-situ technique and in 1993 it was established in the german regulatory for immision surveillance after significant radioactive emissions. For unrestricted release in decommissioning "in-situ gammaspectrometry may be the only method of achieving validation of the release criteria, particularly for large areas outside the buildings" [1].

In a late phase of the decommissioning of a nuclear power plant all components containing a significant inventory of radioactivity are removed, leaving large surfaces with often poorly known contamination levels. Taking in account only buildings of restricted areas in the next 50 years in Germany 7E3 Mg activated and 5E6 Mg contaminated concrete must be released from facilities under decommissioning. Before the ground and the building structures of the facility can be conventionally pulled down, the remaining radioactivity must be determined in order to check the radiological relevance of the concerned part of the plant and to decide the possible path of material release. The basis of assessment may be the so-called "10 μ Sv-concept" [2].

Today the mainly used strategies are analysing samples, taken from the surface or measuring the surface activity via large proportional counters. In both cases we meet severe systematic problems.

1) A commercial contamination monitor meets a minimum detectable activity of 0,1 Bq/cm² Co 60 or Cs 137 on a surface two to ten-times faster than a collimied in-situ spectrometer. On the other hand shielding layers coming from decontamination coatings or migration of the radionuclides in the surface lead to relative small errors performing in-situ measurements whereas contamination monitors are completely inappropriate in the most cases to determine the remaining activity.

2) Taking and analysing samples leads for the single sample to more exact results than in-situ measurements caused by the not completely known source geometry and the measuring time which is in the latter case normally much shorter because of the large number of measurements.

The main problem of the analysis of samples in the laboratory is the estimation of the representativity of a collective of samples. Comparing the results of in-situ measurements and laboratory analysis we noticed substancial differences which could be attributed to strong inhomogenities in the spatial distribution of activity. These were not overcome by sampling.

The importance of this problem can also be shown theoretically. Ferguson [3] determined the number of samples to meet a defined statistical significance for finding an existing contamination. As an example ~200 samples are necessary to find a circular contamination with a diameter of 35 cm on a relative small area of 10 m² with a safety of 95%.

¹ This work was financially supported by an international EC R&D-contract ("Measurement of Spatial Distribution of Contamination on Concrete Surfaces and Build-up of Dusts for the Improvement of Scenarios for Material Release" / CEC Contract FI2D-CT94-0086), which is coordinated by us.

Errors arising from this sort of problems do not appear by measuring with a collimated high-purity germanium detector directly in the regions of interest ("in-situ"). There are only two problems left.

- 1) The vertical distribution of a contamination may be not known well enough. This fact creates a uncertainty which can reach a factor of two. On the other hand this systematic error may be solved and is furthermore small, compared to errors which can affect the quality of the results of traditional techniques.
- 2) A formal problem e.g. in the german radiation protection ordinance is, that the value of a surface contamination has to be achieved by a measurement averaging over 100 cm². Such an averaging area is obviously not adequate for clearance measurements of thousands of square meters of probably contaminated walls, floors, roofs or soils. At the moment this subject is discussed in Germany.

Apart from this the collimated in-situ gamma spectrometry allows to get very fast informations about the radiological state of large areas and -if one takes in account the high quality of these informations- for low costs.

DEVELOPMENT OF AN COLLIMATED IN-SITU SPECTROMETER

Despite the strong worsening effect to the minimal detection limits, it was obvious that for using such a device for clearance measurements it had to be collimated to reduce the field of view. Reasons therefore are that

- possible contaminations must be locateable,
- there must be a well defined averaging area (e.g. some m²), and
- contaminations outside the measured area must be effectively suppressed.

So we designed and constructed a prototype of a high-resolution in-situ spectrometer on the basis of 41% p-type HPGe-detector, surrounded by an optimised collimator made out of brass and a low natural activity lead-bismuth-tin-alloy. As a compromise between portability and shielding effect a attenuation mass per area of ~53 g/cm² was realised for radiation which reaches the collimator in an angle of incidence parallel to the investigated surface. Furthermore the detector is shielded for radiation from the rear hemisphere. Depending on the collimator used and the height of the detector above the surface our spectrometer averages over areas between 0.4 and more than 10 m² in one measurement.

To get quantitative values of contaminations the system had to be calibrated. This needs much more work than for gamma spectrometers in laboratory, where reference sources with relative small volumes for a reduced set of geometries and chemical compositions exist. Performing in-situ gamma spectrometry one has to deal with large sources and any horizontal and vertical activity distributions. They can only be simulated numerically or experimentally. We use three approaches to verify the calibration factors between the full-energy-peak count rate and the specific activity level;

- a modified standard method for uncollimated systems suggested by Beck et al [4] in 1972,
- an experimental calibration by recording a great number of spectra with small sources in different positions and superimposing them as a function of the source geometry and
- a complete numerical simulation modelling the source and the entire detector-collimator-system

All commonly used yield curves with calibration factors based on the standard method have already been verified experimentally. According to our current experience, the two yield curves generally do not deviate from one another by more than 10% in the region of higher photon energy (662 keV and above). Differences of more than 30% have not occurred below 186 keV. These tests make the individual calibrations very reliable, as the two methods are in no way related to one another and the occurrence of identical systematic errors is thus ruled out.

PERFORMANCE OF THE SPECTROMETER

It could be shown that our calibrated prototype is able to meet the maximal activity limits for unrestricted release in Germany in some minutes of measuring time, if the proportionate activity of a strong gamma emitter like Co 60 or Cs 137 is sufficiently high in the nuclide vector. Examples for minimal detectable activities are:

- Under rigid conditions (maximum collimation, some migration of the nuclides in concrete) and normal dose rates due to natural nuclides detection limits of 800 Bq/m² for Co 60 and 1400 Bq/m² for Cs 137 in three minutes are reached.

==> A contamination with a nuclide vector containing only 20-30% of these nuclides can be detected with the requirements of the German radiation protection ordinance.

- For the relatively difficult to measure activity of U 238 (via Pa 234m) a detection limit of 150 Bq/kg is reached after 15 minutes.

==> Uranium can be measured on piles with any enrichment.

==> The observance of exemption values for deposition of waste contaminated with the more difficult to measure nuclides of the nuclear fuel cycle can be proved (f.e. [5])

- In the same time a surface contamination of 500 Bq/m² U 235 is detectable and can be discriminated from the natural Ra 226 and U 235 186 keV background in concrete containing 1 Bq/kg U 235.

Furthermore disturbing radiation from outside the interesting field of view is strongly suppressed. For low and medium energy gamma radiation (<700 keV) from nuclides distributed vertically and horizontally homogeneous in soil 90% of the total photon flux appear from an angle of $\pm 48^\circ$ relative to the detector axis in the collimated case. Without collimator this value would rise to 82° , so it would be impossible to localise a source.

APPLICATION OF THE SPECTROMETER

Aside from the development of such a spectrometer we made experiences with our prototype inside the restricted area and outside the buildings of several nuclear power plants under decommissioning like KKN, KRB-A and VAK in Germany and G 3 and RAPSODIE in France by painting a grid over the area of interest and measuring at each grid point. Measurement campaigns in facilities of the nuclear fuel cycle were performed in NUKEM-A, HOBEG and WAK in Germany.

Up to now with this campaigns new knowledge about the lateral distribution of contaminations, the variability of its composition or -by comparing with other measuring techniques- their systematic uncertainties could be quantified. The description of the results of these campaigns would exceed the scope of this compact. As examples we

- mapped the short distance variability of the Chernobyl Cs 137 fall-out by scanning 1500 m² grassland completely with 3 m² surface area per single measurement,
- showed for a contaminated floor for one room in the restricted area of a nuclear power plant that the Co 60 / Cs 137 ratio was completely indefinite (variation over more than three orders of magnitude) and
- investigated large deviations in the measured surface activity via proportional counter and in-situ for several surfaces with different histories and contamination scenarios.

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