

Mobile Nuclear Laboratory for In-Situ Measurements in NPPs

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Abstract. In-situ gamma- and alpha-spectrometry, beta- and neutron counting and gamma-dosimetry are utilized in Paks Nuclear Power Plant to assist normal maintenance operations as well as elimination of malfunctions with their consequences. Devices, methods applied and results, experiences achieved during the last 25 years are described. They are well applicable for the new generations of the future reactors as important additional safety measures.

1. INTRODUCTION

The Laboratory for Nuclear Safety and Techniques was established by the University of Debrecen, Hungary and Quantechnologies Research and Development Co in 2005 to utilize the results of the basic research achieved earlier in the investigations of nuclear fission, gamma-spectrometry and related nuclear physics fields as well as the two decade experience on the in-situ gamma-spectrometry at Paks NPP.

Gamma-spectrometry of the primary loop pipes, ion exchange columns and steam generators is utilized regularly to all the 4 units of VVER-440/213 reactors at Paks NPP since 1985. (Three similar measurements were also carried out in Biblis-A, Germany.) The isotope selective, non-destructive analysis is applied to determine the activity of corrosion/erosion products and fission nuclides. The measurements are carried out after the yearly shut-down with a cooling time ranging from several days to three weeks. Detected nuclides are: ⁵¹Cr, ⁵⁴Mn, ⁵⁸Co, ⁵⁹Fe, ⁶⁰Co, ⁶⁵Zn, ⁹⁵Zr, ⁹⁵Nb, ^{110m}Ag, ^{122,124,125}Sb, ¹⁰³Ru, ¹⁰⁶Ru/Rh, ¹³¹I, ^{134,136,137}Cs, ¹⁴⁰Ba/La, ¹⁴⁴Ce/Pr, ¹⁴⁸Pm, ¹⁵⁴Eu. Fuel element investigations were also performed.

Alpha-spectrometry is utilized to investigate transuranium contamination on surfaces being in contact with the primary loop moderator. Neutron counting is devoted to the detection of transuranium isotopes in locations non-accessible by alpha-detectors. Beta-detection (as well as alpha- and gamma-spectrometry) is utilized for the measurement of non-fixed surface contamination by sample taking.

2. EXPERIMENTAL

All the measurements are carried out at the premises of the power plants requiring mobile detectors, sophisticated electronics and data handling/reduction systems.

2.1. Gamma-spectrometry

Lead-shielded and collimated, liquide nitrogen cooled HpGe-detectors with different sensitive volumes of 1, 3, 10 and 100 cm³ and a clover type diode of 4x100 cm³ (with add-in mode) are used for high-resolution, high-sensitivity gamma-spectrometry. Normal temperature CdZnTe and usual NaI(Tl) scintillation detectors are also applied in special circumstances. Absolute full-energy peak efficiency

for the diverse geometry (extended) sources are determined by experiments and calculations to produce final results as surface (or volume) activities in Bq/m² (or Bq/lit). Validation was performed by comparing the results with those of radiochemical samples taken from the same surfaces, model experiments and simulations.

Electronics, data acquisition and evaluation are characterized by remote operation (cable length of 100 m), high counting rate (up to 200 kcps), PC-controlled digital signal processors. Results of the primary side measurements regard mainly the water chemistry and associated effects of the operation by the corrosion/erosion nuclide activities and their long term behavior. Fission product activities characterize the fuel-element hermeticity.

Investigated locations of the regular yearly measurements in the primary side include: 2-2 points at the hot-legs and cold-legs of each of the 6 loops (see Fig.1), 8-10 heights on the ion exchange columns (anion, cation and mixed-bed), 16-17 spots along the steam generator axis. Gamma-dose profiles are also determined.

Regular in-situ gamma-spectrometry and its results for the water chemistry in the primary circuits were published in details elsewhere [1, 2].

2.2. Alpha-spectrometry

In-situ alpha-spectrometry for the surfaces of fuel assemblies and technical pool were performed to investigate transuranium contamination. These surfaces are rather rough and sometimes covered by inhomogenous layers. Thus the spectra are distorted although vacuum pump is utilized to decrease the energy loss and straggling of the alpha-particles (and fix the detector). Passivated implanted planar Si (PIPS) diodes are utilized the surface of which is cleanable [3]. Contamination from ^{238,239,240}Pu, ²⁴¹Am and ^{242,244}Cm were determined [4].

2.3. Neutron-detection

Neutron signatures from spontaneously fissioning transuranium isotopes are measured by ³He proportional chambers. Plastic slab moderators made the efficiency higher. This technique is hard to give absolute isotope amounts therefore it was used as a warning message for the presence of heavy nuclides, mainly ^{242,244}Cm.

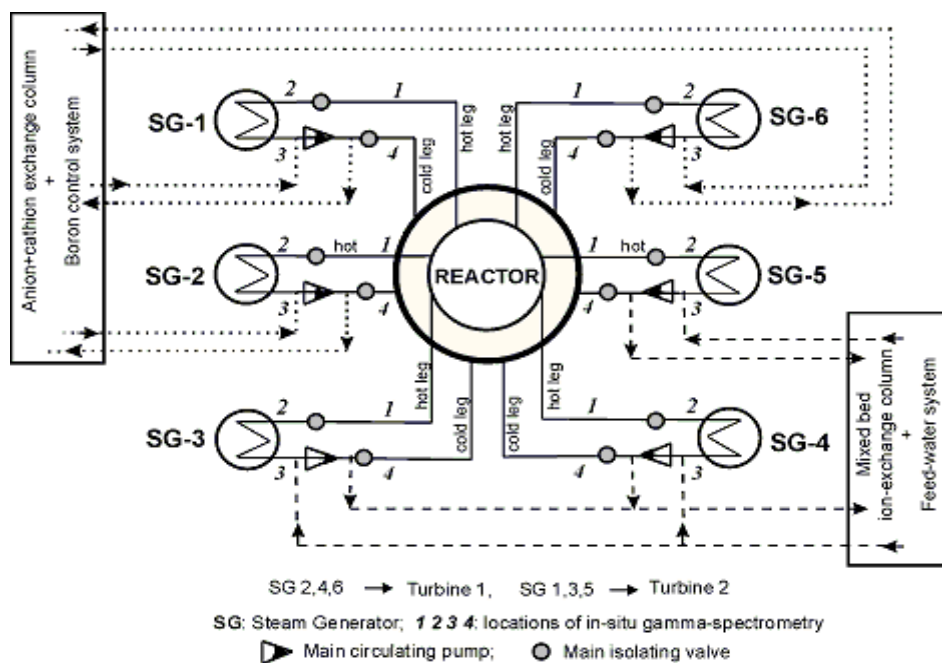
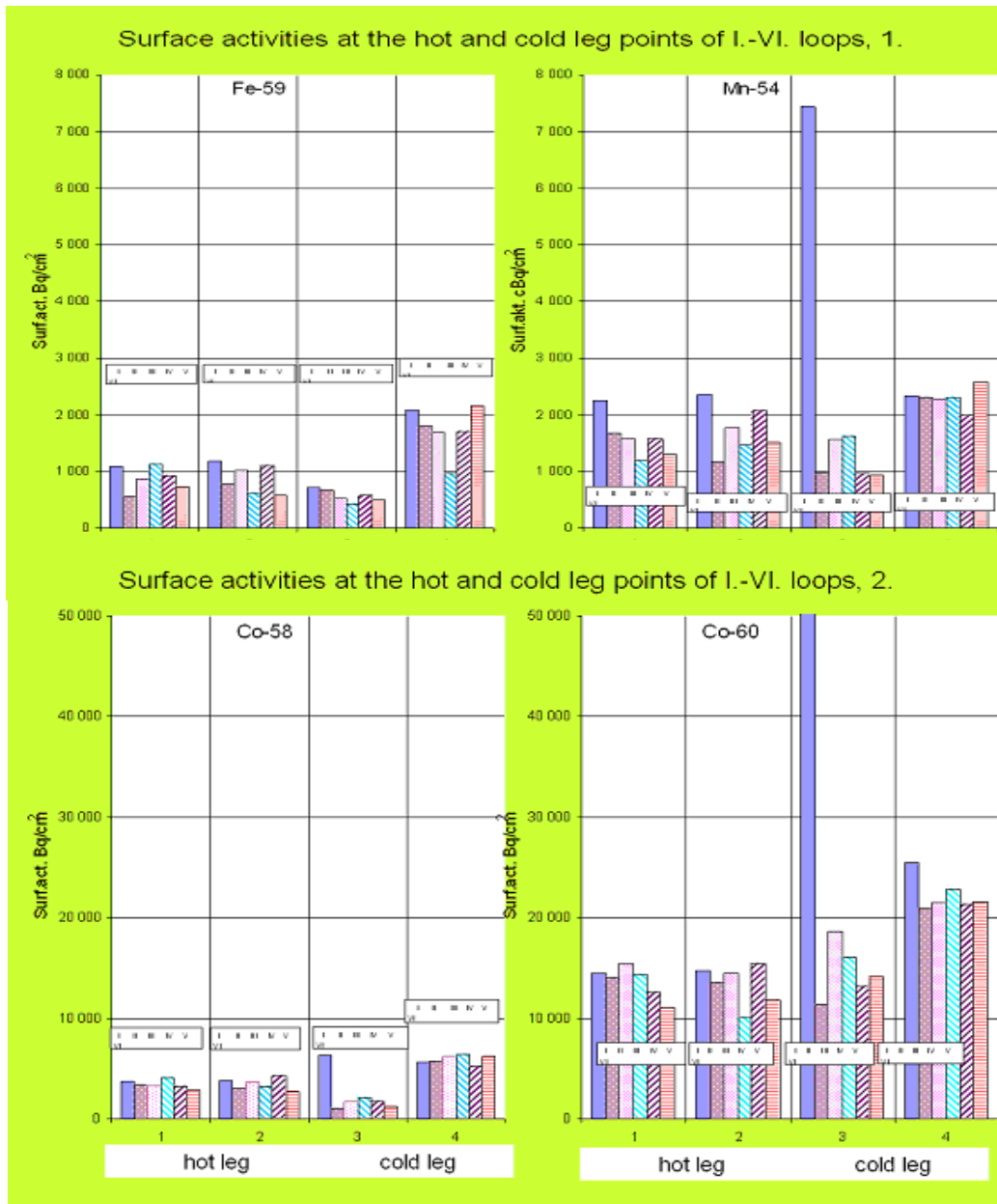


Fig. 1. Scheme of the VVER-440/213 unit with in-situ assay points 1 – 4 on the 6 loops



2.4. Beta-counting

This technique is applied to measure activity of rubbed samples taken from contaminated surfaces. Two computer controlled, pancake shaped GM-tubes are generally utilized the counts of which are summed to increase the solid angle (detection efficiency).

3. RESULTS AND DATA ANALYSIS

An invaluable, detailed dataset of in-situ gamma-spectrometry and moderator activity was established during the ~100 reactor-years the analysis of which is in progress.

3.1. General characteristics of the results

Some short-term and long-term observations were attained for the main corrosion products: differences and their time variations among the surface activities of the cold-legs and hot-legs reflecting some ageing behavior of the inner layers; effects of the auxiliary device operation like boron control circuit, deaeration of tap water supply, saturation of the ion exchangers; lost material; corrosion transport. Inspection of the decontamination process in steam generators, its time evolution and isotope selective efficiency were also investigated.

Information attained from the measurements can be summarized as follows [5]:

- a) Analysis of one reactor unit in one cycle: actual corrosion state is observed,
spatial distributions: ion exchanger (resin saturation), steam generator;
differences among legs/points (local failure), lost material.
- b) One reactor unit, many cycles: case a) + changes in the water chemistry
evolution of the distributions: material transport, ageing
forecasts for the contamination, dose situation
life-time prolongation, decommissioning
correlations among parameters of water chemistry, reactor operation.
- c) Many reactor units, one cycle: case a)
+ differences in the water chemistry, material composition.
- d) Many reactor units, many cycles: a) + b) + c)
+ ageing differences, water chemistry, variations in the ageing.

Some of the main characteristics of the measured surface activities are summarized below for corrosion product behavior at the loop points.

3.2. One-cycle experiences for a reactor unit

Figure 2 shows the surface activity of the main corrosion products measured by in-situ gamma-spectrometry at a given maintenance outage. Different behavior of the ^{59}Fe (characterizing iron transport), ^{54}Mn (iron, too), ^{58}Co (nickel) and ^{60}Co (cobalt) is well observable. Point 3 of Loop I. has extremely high activity for ^{60}Co and ^{54}Mn , less absolute (but relatively strong) contamination is shown by ^{58}Co , while ^{59}Fe is not affected at all. Considering the same origin of ^{59}Fe and ^{54}Mn from target material Fe having very different half-lives the probable time of the contamination process may be estimated. Early release of corrosion products decreased the ^{59}Fe ($T_{1/2}=44.5$ d) activity much more than that of ^{54}Mn (312.2 d).

3.3. Long-term characteristics of a reactor unit

These results are essential in the normal operation and maintenance, forecast of the future contamination and dose levels, as well as for power upgrade and life-time prolongation programs. Theoretical investigations of the corrosion process with a computer code based on a physico-chemical model and its results compared to the measured activities are performed, too.

Cold and hot legs have different contaminations according to their position relative to the reactor vessel which is manifested in temperature variations. Figure 3 shows these effects as a function of time (cycle) for the single measurement points. The cold leg – hot leg surface activities differ by a factor of 4 – 12. There is yet a fine effect: the activity ratio for the cold leg points 3 and 4 is

dependent on the loop positions. The anion and cation ion exchange columns as well as the boron control system are connected to loops I, II and VI while the mixed bed ion exchanger with the feed water system are parts of loops III, IV and V, see Fig.1. Thus the activity sequence in the previous system is $3 < 4$ while that of the latter is opposite as $3 > 4$.

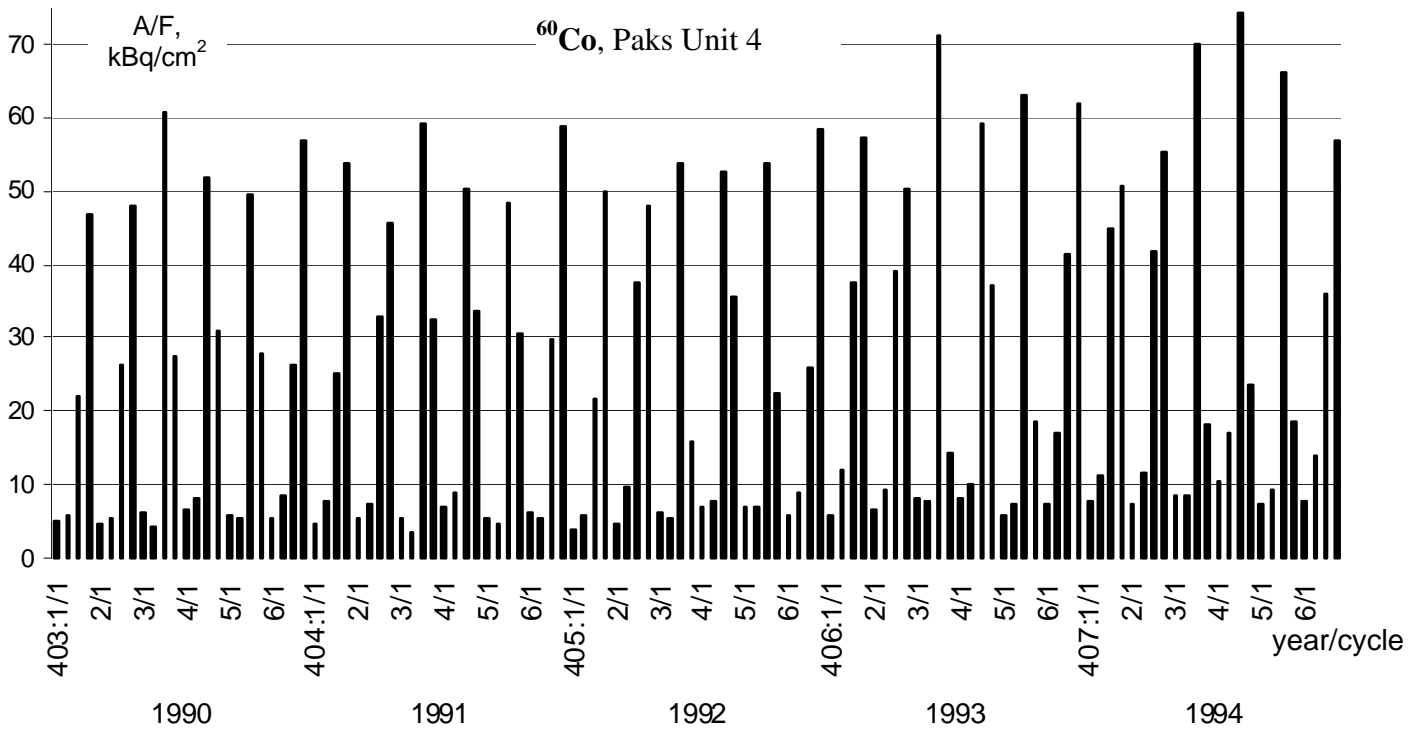


Fig. 3. Surface activity of single points 1, 2, 3 and 4 of loops I – VI for ^{60}Co at Paks Unit 4 (one point of a loop is marked, only: 6/1 means point 1 at loop VI)

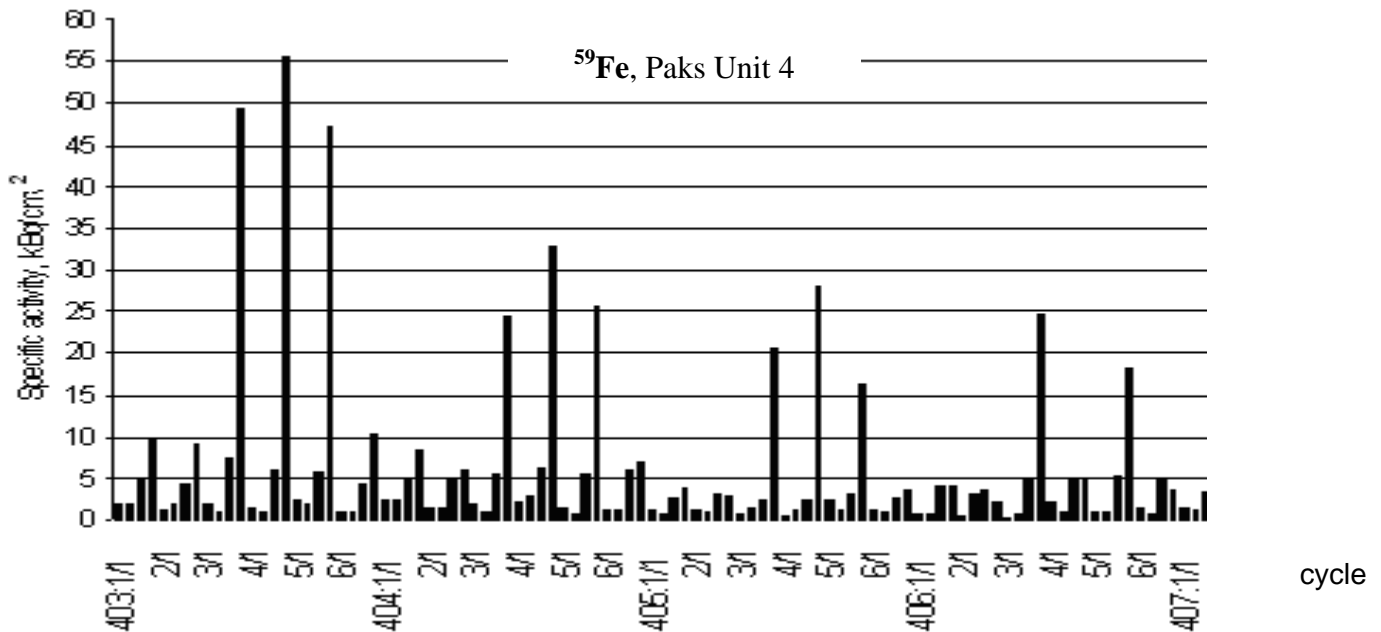


Fig. 4. Surface activity of single points 1 – 4 of loops I – VI for ^{59}Fe at Paks Unit 4.

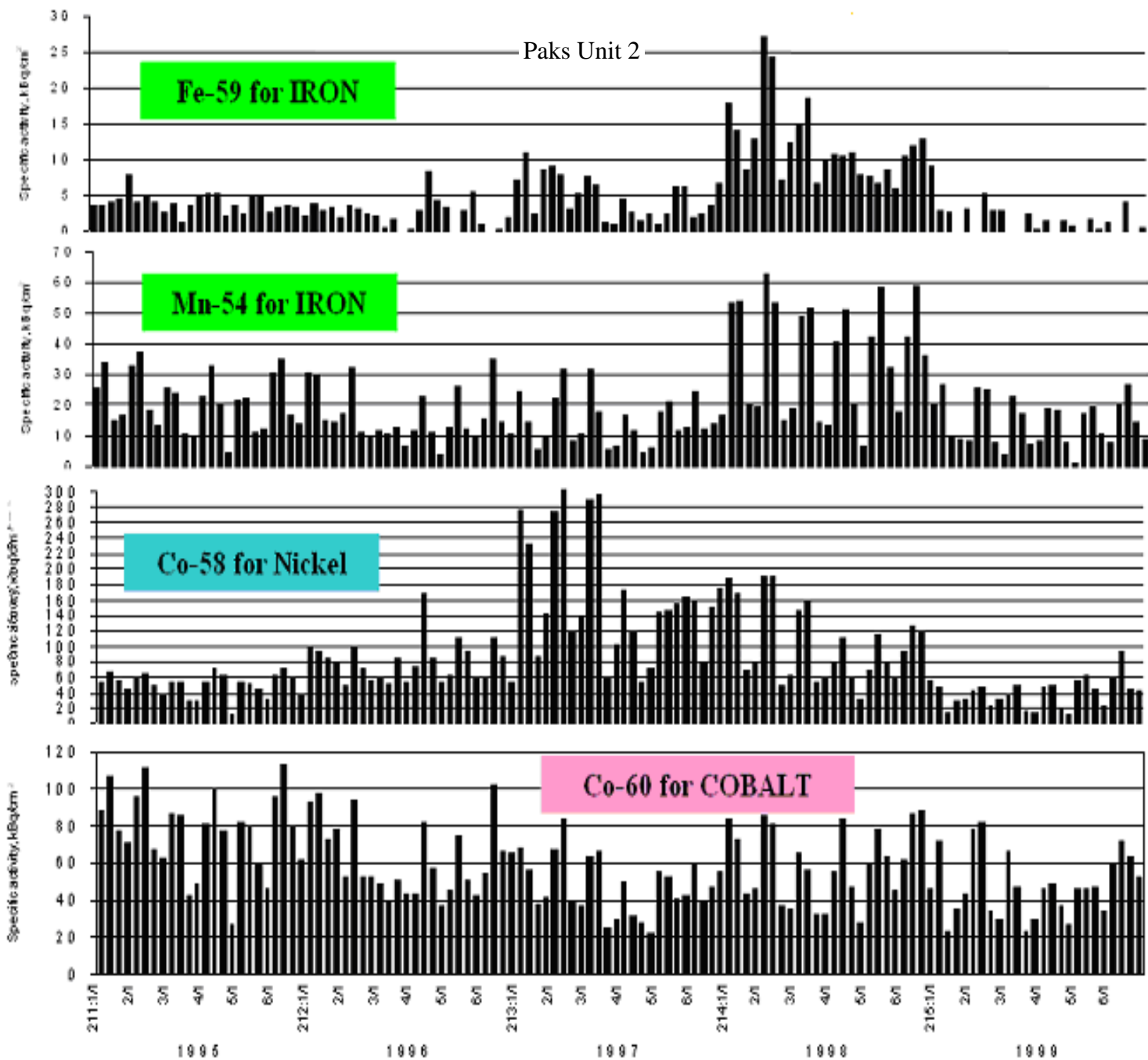


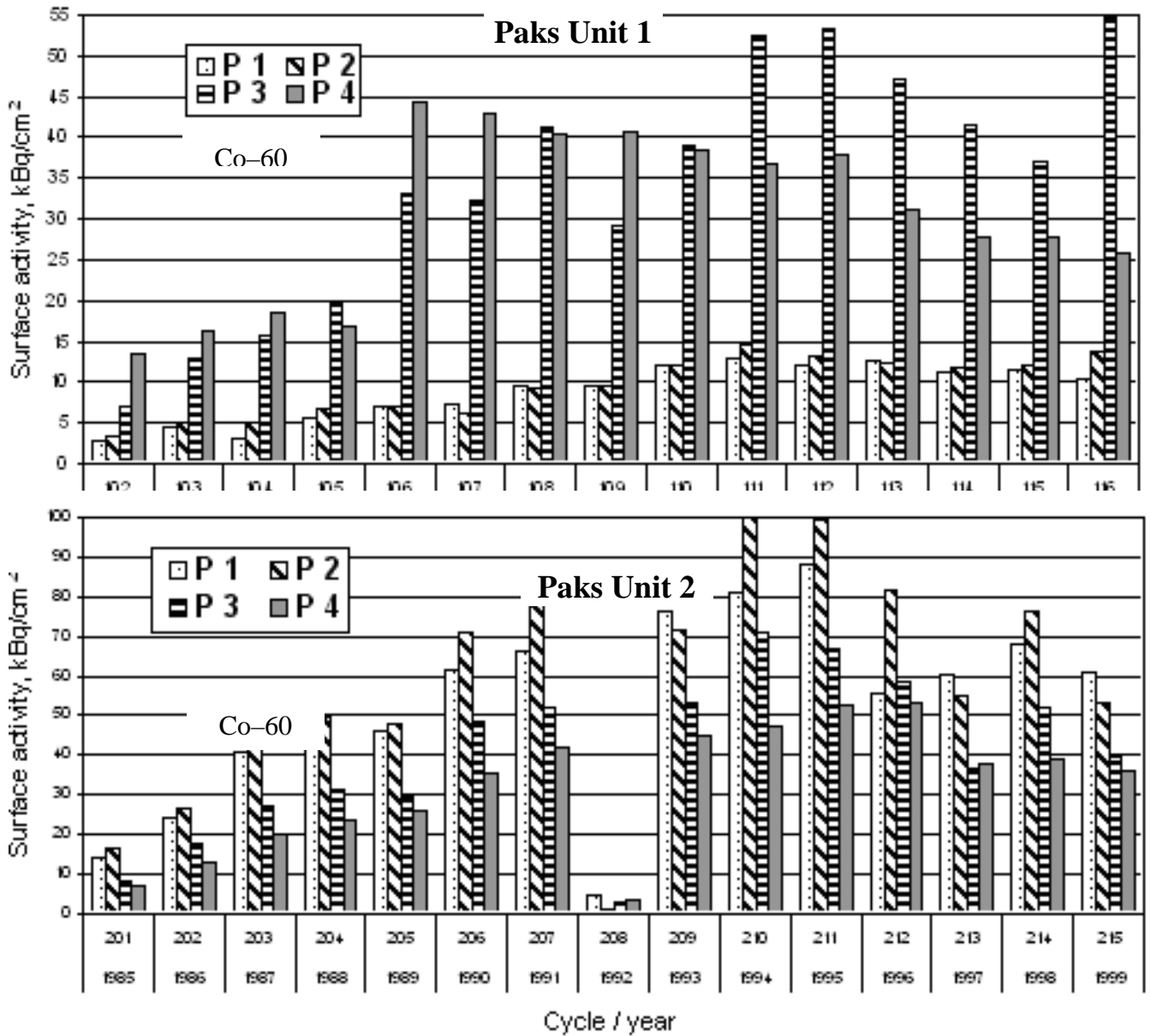
Fig. 5. Surface activity of single points 1 – 4 of loops I – VI for the main corrosion nuclides at Paks Unit 2 for the period of 1995 – 1999.

These points are at the main pump output in the feed water sub-circle. This „extra” corrosion was due to a malfunction of the deaerator of the feed water. Activity of ^{60}Co did not show similar effects. The fault might occur for a short period of time in many cycles.

Different time behavior may be observed in Fig.5 for the corrosion products at Paks Unit 2. Nuclides ^{59}Fe and ^{54}Mn have a general maximum around year 1998 while the peak for ^{58}Co appeared one year before and lasted two cycles. ^{60}Co seems not to be affected much. The common feature for the two Fe-targeted nuclides is obvious, although ^{54}Mn had a more clear hot leg activity enhancement over the cold leg points. It might be caused by the decay of ^{59}Fe by its half-life shorter than that of ^{54}Mn . The origin of this interesting picture was a lost steel sheet broken to pieces, later having been powdered and dissolved into the primary pipes. Its material composition seemed to be different from that of the „normal” tube wall.

3.4. Variations among reactor units

Hot leg – cold leg activity inversion in Paks Unit 2 compared to the „normal” distribution in Unit 1 is clearly seen in Fig.6. Six-loop averaged point activities were calculated to emphasize the behavior of the legs. While cold leg contamination is higher than that of hot leg by a factor of 3–4 at Unit 1 this ratio is inverted in Unit 2 by 1,5–2. This inversion decreased and later changed to „normal” ratio here like in Unit 1. It is interesting to note that with ageing other units also changed their characteristics to and back. A general trend of activity increase is also observable from the data.



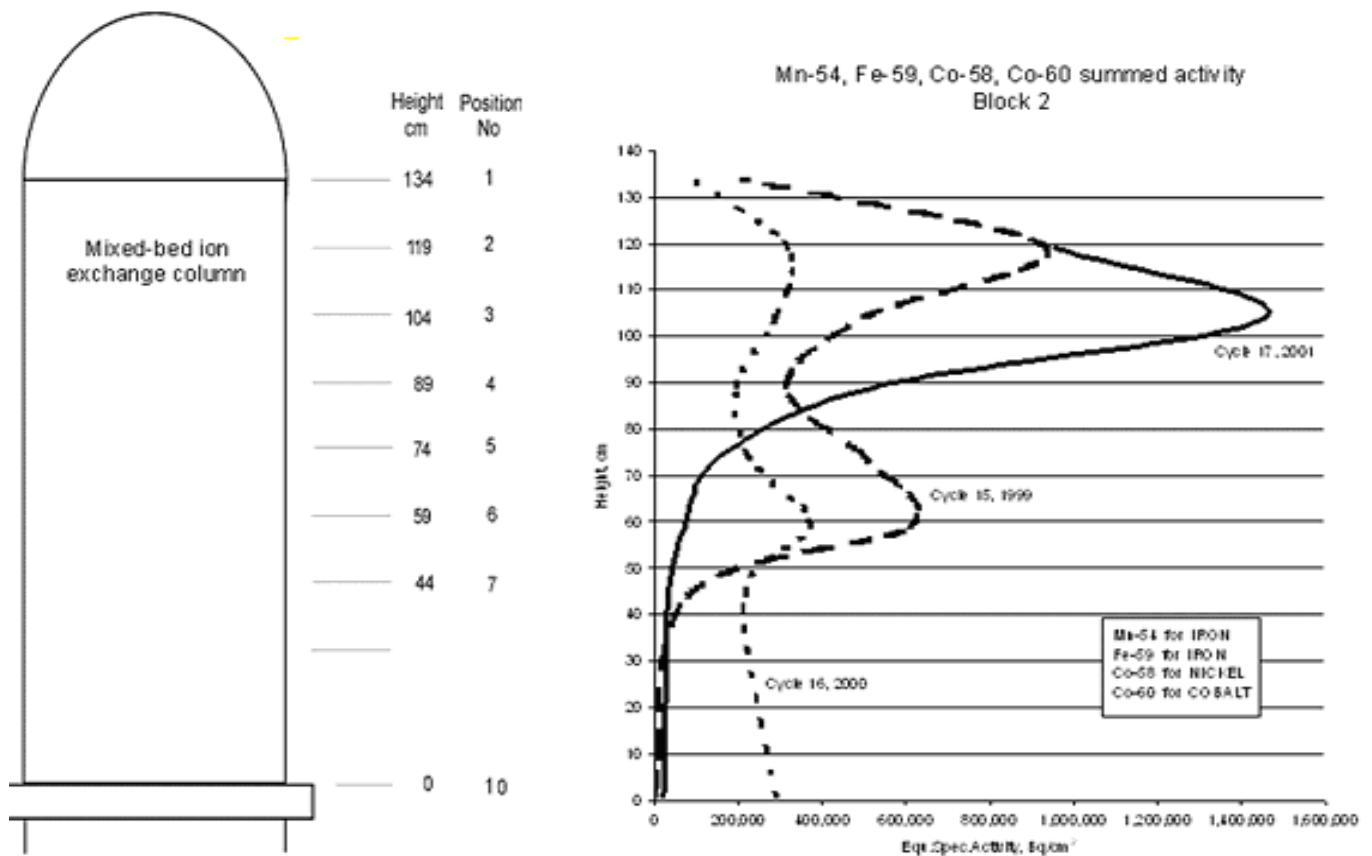


Fig. 7. Spatial distribution of the summed corrosion product surface activity on the mixed bed ion exchange column for three cycles 1999 (---), 2000 (- - -) and 2001 (—) at Unit 2

3.5. Ion exchange columns

Mixed bed ion exchange column is measured mainly. Vertical isotope distribution is determined by in-situ gamma-spectrometry to recognize the saturation of the resin and to control its proper operation. Fig.7. shows such a profile for the corrosion products at different cycles.

3.6. Steam generators

Horizontal isotope map is determined for one device a reactor unit during the maintenance period. Typical results for the main corrosion materials and the steam generator scheme are shown in Fig.8. Measurements are carried out from the hot collector's side. $^{58,60}\text{Co}$, ^{124}Sb (the 3rd highest activity) and ^{54}Mn are dominating the nuclide distribution. There is a well observable geometrical asymmetry in the contamination which may be interpreted on the basis of the inner structure and water flow characteristics.

This method is capable also for monitoring the decontamination procedure of the steam generators. Time dependence of the activity of different isotopes was determined by in-situ gamma-spectrometry. Dissolution, desorption and washing phases might be followed successfully and the efficiency of the full process determined.

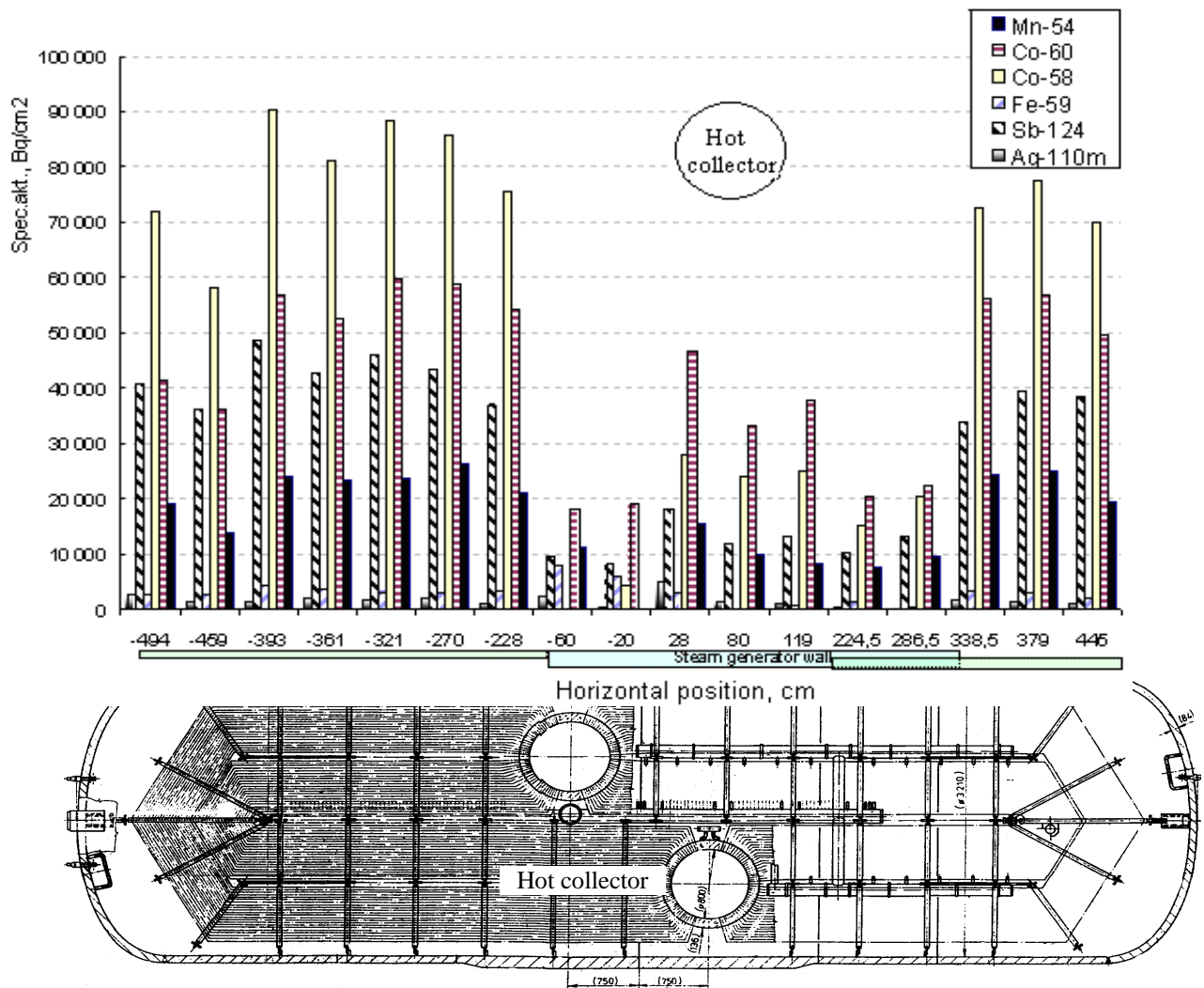


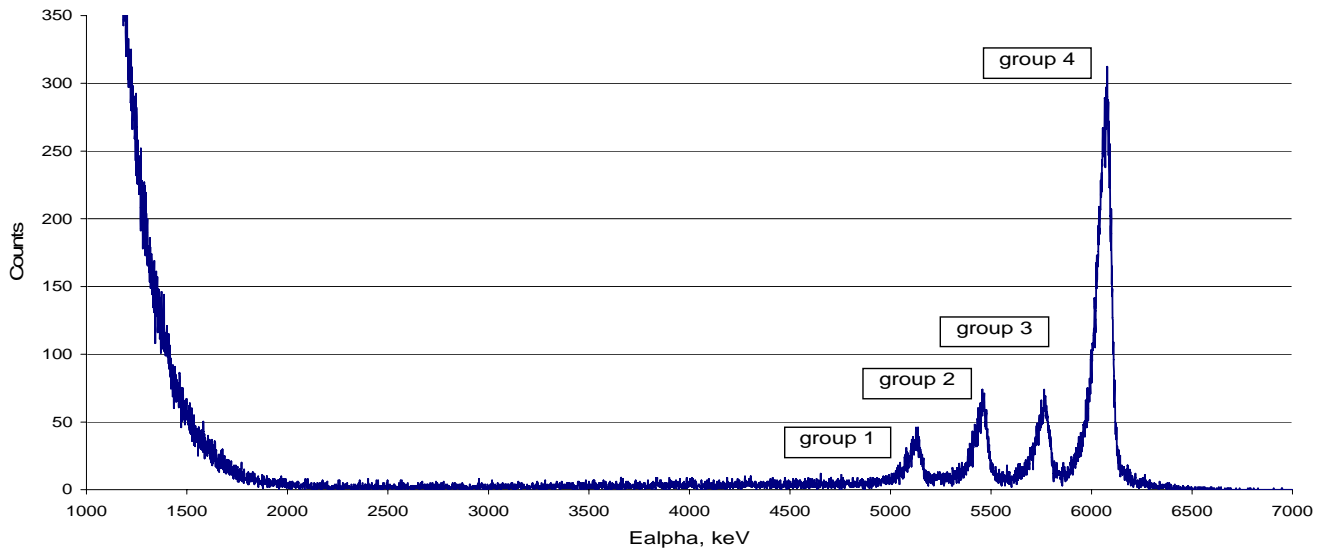
Fig. 8. Spatial distribution of the main contamination nuclides at Steam Generator 5 of Unit 3 in 2009

4. SPECIAL NUCLEAR TECHNIQUES

A malfunction, caused during fuel cassette cleaning, initiated some new nuclear techniques to be introduced. Radioactive contamination for surfaces of fuel assemblies and pools (technical, cooling, fuel transportation, etc) were analyzed by gamma- and alpha-spectrometry. The gamma-spectrometry was performed for the usual targets and for under water surfaces. In the last cases special containers („submarines”) were constructed with remotely controlled variable size collimator-shutter. Spectrometers include some of the aforementioned HpGe detectors as well as CdZnTe-crystals of $1.5 \times 1.5 \times 1 \text{ cm}^3$ sensitive volume. Although the energy resolution of the latter is worse than the formers (but better than that of scintillators), the high temperature operation and the appropriate geometry, small overall size make them extremely valuable for special measurements in hard environmental conditions.

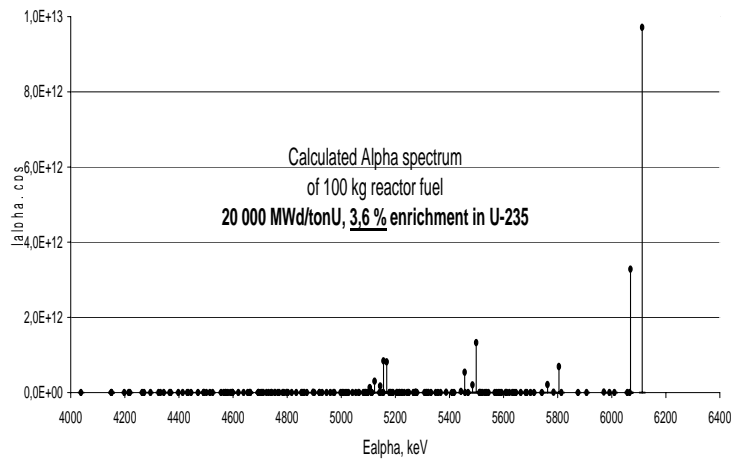
Dry surfaces of the above mentioned pools and assemblies were analyzed by in-situ alpha-spectrometry, too. A cleanable Passivated Implanted Planar Silicon (PIPS) detector housed in a miniature lead shielded chamber was fixed onto the surfaces by vacuum and some mechanical pressure. It measured alpha-spectra for the determination of surface activities from isotopes of $^{238,239,240}\text{Pu}$, ^{241}Am , $^{242,244}\text{Cm}$. Typical alpha-spectra with calculated ones are shown in Fig.9. They refer to contaminated surfaces of fresh fuel cassettes picked-up from the cooling pond. Experimental correlations among rare-earth fission products measured by gamma-spectrometry and transuranium nuclides determined from alpha-spectra were established to make the latter's activity to be estimated for non-attainable locations (eg. inner wall of steam generator heat transfer tubes). Similar correlations with spontaneous fission neutron activity were also applied to observe transuranium (mainly Cm) isotopes from "buried" sources.

Measured alpha-spectrum
Casette 136 60506, position 3: Zr-neck



*alpha-spectra for reactor fuel
contaminating cassette surfaces*

- Group1: 5.1 MeV $^{239,240}\text{Pu}$;
- Group2: 5.5 MeV ^{238}Pu , ^{241}Am ;
- Group3: 5.8 MeV ^{244}Cm ;
- Group4: 6.1 MeV ^{242}Cm .



All these techniques and the appropriate methods are used for the analysis of activity waste wells of nuclear power plants, waste deployment sites, general nuclear industry, medical nuclear centers, environmental radioactivity (NORM, TENORM analysis).

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