RADIONUCLIDE EXPRESS ANALYSIS FOR THE CONTROL OF NUCLEAR MATERIALS AND ISOTOPE SOURCES

N.V. Ivashkin

Russian Federal Nuclear Center - Zababakhin All-Russia Research Institute of Technical Physics (RFNC-VNIITF), Snezhinsk, Russia

e-mail: n.v.ivashkin@vniitf.ru

Content

1 Introduction: control-related problems and radionuclide express analysis.

1.1. The purpose of the control and the intended use.

The general purpose of the control is to provide ecological safety (in terms of nuclear and radiation safety) of the staff, population and environment under the use of nuclear energy.

The areas covered by the control should be as follows:

- **Nuclear fuel cycle (NFC) and nuclear weapons complex (NWC);**
- **Legal and illegal handling, turnover, and export/import of NM and isotope sources;**
- **Testing of nuclear explosive devices; and**
- **Surveillance of areas free from nuclear weapons.**

1.2. Urgency of improving the technologies of production, using and handling nuclear materials and isotope sources is conditioned by:

- **Suppositions of nuclear proliferation (de-facto emergence of new nuclear states);**
- **Enhancement of radiological terrorism threat under the circumstances of nuclear technology development and, notably, of electrical power engineering in developing states;**
- **Potential threat from the part of radiation emergency accidents in nuclear sites of NFC and NWC whose number has been continuously increasing;**
- **Expansion of medical use of radioactive isotopes and development of nuclear medicine.**

1.3 Features of radionuclide express analysis as a technical segment of the control system:

- **Acquiring direct data on the presence, isotope composition and characteristics of radiation from the objects and sites under control;**
- **Possibility to provide on-site inspection through measurements using non-intrusive methods and technical means (including portable ones) on mobile carriers;**
- **Lower cost and higher efficiency of measurements;**
- **Non-distructive on-site analysis of samples and object surveillance using remote gamma-spectrometry;**
- **Use of instrumental methods of alpha- and gamma-spectrometry, and beta-radiometry of samples (without using radiochemistry).**

1.4 Monitored materials, radionuclides and high-activity isotope sources:

1. Source and special fissionable material *(as defined in Article XX of the Statute of the IAEA)*:

1.1. Source material: Natural U *(0,72 % ^{235}U)*, **Depleted U** ***(0,2-0,3%*** *^{235}U)*, **Th;**
1.2. Special fissionable material: ^{239}Pu *(≤ 80% ^{238}Pu)*, **^{233}U, ^{235}U** *(≥20% ^{235}U)*

2. Nuclides *(RF Trigger List)*: **^{237}Np, ^{241}Am, ^{243}Am, ^{252}Cf.**

3. Isotope Sources:

- **Industrial irradiators** (^{60}Co, ^{137}Cs, ^{192}Ir);
- **Radioisotopic generators** (RIG: ^{90}Sr, ^{210}Po, ^{238}Pu);
- **Radiotherapy** (**γ-Knife**: ^{60}Co, ^{137}Cs, ^{192}Ir);
- **Well-Logging** (^{241}AmBe, ^{239}PuBe, ^{226}RaBe, ^{252}Cf);
- **X-Fluorescent analyzers. Nuclear Medicine Sources** (therapy, tomography).

2. Radiation characteristics of nuclear materials and radionuclides

Table 1 – Characteristics of alpha and quantum-emissions of nuclear materials and radionuclides ***

Isotopes	Alpha-emissions		Gamma-emissions		X_K (X_L)-emissions	
	Energy, keV	Intensity (α/dis.), %	Energy, keV	Intensity (γ/dis.),%	Energy, keV	Intensity (γ/dis.)
^{233}U	4823/4779	83/15	42.4/164.5	$8.6\cdot10^{-2}/6.6\cdot10^{-3}$	93.3; (15,7*)	$1.2\cdot10^{-2}$; (5.3**)
^{234}U	4768/4717	72/28	53.2/120.9	$1.2\cdot10^{-1}/3,4\cdot10^{-2}$	93.3; (15,3*)	$4.4\cdot10^{-3}$; (10.2**)
^{235}U	4370/4354	25/35	143.8/185,7	10.96/57.2	89.9/93.3; (15,3*)	3.56/5.76; (22**)
^{238}U	4195/4145	77/23	63 (^{234}Th)/1002 (^{234m}Pa)	$4.8/8.4\cdot10^{-1}$	93.3; (15,3*)	$1.8\cdot10^{-3}$; (7.94**)
^{237}Np	4788/4771	46/20	86,5/143.2	$12,4/4,3\cdot10^{-1}$	95.9; (13,29)	2.9; (18.7)
^{238}Pu	5499/5456	71/29	43.5/99.8	$4.0\cdot10^{-2}/7,0\cdot10^{-3}$	94.6/98.4; (13,56)	$1.0\cdot10^{-4}/1.7\cdot10^{-4}$;
^{239}Pu	5157/5144	73/15	375.0/646.0	$1.56\cdot10^{-3}/1,5\cdot10^{-5}$	94.6/98,4; (13,58)	$6.9\cdot10^{-3}$; (1.5)
^{240}Pu	5168/5124	73/27	104,2/642.3	$7.1\cdot10^{-3}/1,4\cdot10^{-5}$	98.4; (13,58)	$1.0\cdot10^{-4}$; (3.6)
^{241}Pu	4896	$2.0\cdot10^{-3}$	103.7/148.6	$1.0\cdot10^{-4}/1.9\cdot10^{-5}$	100.5; (16,0)	$*1.0\cdot10^{-3}$; $**(7\cdot10^{-4})$
^{241}Am	5486/5443	85/13	59.5/662,0	$35.9/3.6\cdot10^{-4}$	101.1; (17,8)	$1.9\cdot10^{-3}$; (20.0)
^{252}Cf	6118/6075	81.7/15,1	43.4/100.2	$1.5\cdot10^{-2}/1.2\cdot10^{-2}$	109.3; (18*)	$4.0\cdot10^{-5}$; (6.1**)
* Average Energy $<E_{XL}>$; **Sum Intensity $\sum I_{XL}$						

****Чечев В.П., Кузьменко Н.К., Сергеев В.О., Артамонова К.П. Оцененные значения ядерно-физических характеристик трансурановых радионуклидов.− Справочник. М.: Энергоатомиздат, 1988.*

Table 2 – Specific activity of isotopes; neutron yields of spontaneous fission and (α, n) - reactions*.

Isotope	Specific Activity, Bq/g	Specific Yield, (n/s·g)	Chemical Compound	Specific Yield, (n/s·g)
^{233}U	$3.6 \cdot 10^{8}$	$4.3 \cdot 10^{-4}$	$^{234}UF_6$	$3.9 \cdot 10^{2}$
^{234}U	$2.3 \cdot 10^{8}$	$7.8 \cdot 10^{-3}$	UF_6 (90% ^{235}U)	3,4
^{235}U	$8.0 \cdot 10^{4}$	$1.1 \cdot 10^{-5}$	UO_2 (90% ^{235}U)	2.3 (calculation)
^{238}U	$1.2 \cdot 10^{4}$	$1.4 \cdot 10^{-2}$	$^{238}UF_6$	10.1
^{237}Np	$2.6 \cdot 10^{7}$	$1.2 \cdot 10^{-4}$	UO_2 (4,4 % ^{235}U)	$1.2 \cdot 10^{-2}$ (calculation)
^{238}Pu	$6.3 \cdot 10^{11}$	$2.6 \cdot 10^{3}$	$^{238}PuO_2$	$1.3 \cdot 10^{4}$
^{239}Pu	$2.3 \cdot 10^{9}$	$2.2 \cdot 10^{-2}$	$^{239}PuO_2$	40 (расчет)
^{240}Pu	$8.4 \cdot 10^{9}$	$1.0 \cdot 10^{3}$	$^{239}PuF_4$	$4.6 \cdot 10^{3}$
^{241}Am	$1.3 \cdot 10^{11}$	1,2	$^{241}AmO_2$	$2.5 \cdot 10^{3}$
^{252}Cf	$2.0 \cdot 10^{13}$	$2.3 \cdot 10^{12}$	$^{239}PuBe_{13}$	$5.7 \cdot 10^{4}$

** Ф.Ф. Фролов. Ядерно-физические методы контроля делящихся веществ. М.: Энергоатомиздат, 1989, с.177.*

Table 3 – Controlled quantity of nuclear materials and high-activity isotope sources. Radiological impacts of the heaviest accidents.

Materials/ Nuclides	Generation Process	Estimated Quantities
WG Pu оружейного качества	Uranium reactor irradiation	➢ **~10^{18} Bq**; ~ 250 t; thousand units of storage
Pu, spent fuel	Recovery in NPP fuel	➢ **~6 10^{18} Bq**; ~ 2000t; hundred units; stockpiling ~100 t/year
Pu, dedicated from NPP spent fuel	Radiochemical (RC) extraction	➢ ~**6 10^{17}** Bq; ~ 200 t; hundred units of storage
WG U	Isotope enrichment	➢ ~3·10^{15} Bq; ~ 2000 t; thousand units
DU	Uranium enrichment waste	➢ ~10^{16} Bq; 10^{5} t; thousand units of storage
Tritium	Reactor irradiation	➢ ~100 kg (specific activity– 3,6·10^{17} Bq/kg)
Radioisotope sources (^{60}Co, ^{137}Cs, ^{192}Ir, ^{90}Sr, ^{238}Pu)	Reactor irradiation, RC-extraction from spent fuel	➢ **~10^{19} Bq**; max activity of one unit–~4·10^{17} Bq (^{90}Sr in RIG); thousand units
γ-sources for Nuclear Medicine (^{60}Co, ^{137}Cs, ^{125,131}I…)	Reactor and cyclotrons irradiation, RC- extraction	➢ max activity of one unit ~10^{14} Bq (^{131}I); thousand units
Spent Fuel	Reactor irradiation	➢ **2 10^{19} Bq**; ~2 10^{13} Bq/kg (3-year expose spent fuel)
RC Plant «Mayak» (USSR), 1957	Explosion of HLW tank	❖ **7.4·10^{16}** (Σ); 2.0·10^{15} (^{90}Sr)
Windscale (GB), 1957	Fuel melting (Pu production reactor)	❖ 7.4·10^{14} (^{131}I); 2.2 10^{13} (^{137}Cs)
NPP “Three Mile Island” (USA), 1979	Fire, melting of reactor core	❖ 5 10^{11} (^{131}I); 1.5 10^{10} (^{137}Cs)
Chernoble NPP, (USSR), 1986	Melting of reactor core, power unite explosion	❖ **~2·10^{18}** (Σ); 6.4·10^{16} (^{137}Cs)
NPP «Fukushima-1» (Japan), 2011		❖ **~1.5·10^{17}** (^{131}I); 1.2·10^{16} (^{137}Cs)
➢ Estimation of worldwide quantity; Σ –Summing activity in accidents (Bq)		

3. Methods and equipment used for express analysis

3.1 Method characteristics

Main methods:

- On-site remote γ-spectrometry of objects from various type carriers;
- Sampling and on-site γ, α–spectrometry in mobile field laboratories; and
- Non-destructive γ-spectrometry analysis and neutron measurements of nuclear materials.

MDA – minimum detectible activity;

$$MDA = [2.71 + 4.66 (S_{фп})^{1/2}] / (\tau \cdot m \cdot n_{\gamma} \cdot \varepsilon_{ф}), \quad (3)$$

$S_{фп}$ – "pedestal" square under photo peak" (pulse); τ- exposition time (s); m – mass of sample (kg); n_{γ} - quantum yield of gamma-line (γ/dis.);

Table 4 –Relative (with respect to analytical methods) sensitivity of gamma-monitoring methods applied to environmental objects.

Nuclide	Soil		Surface fallout		Direct γ-spectrometry of samples	
	Aero γ-survey	Above-ground γ-survey	Aero γ-survey	Above-ground γ-survey	Soil, sediments	Water, solution
^{137}Cs	$2 \cdot 10^{-2}$	0,3	~ 10^{-3}	$1 \cdot 10^{-3}$	0,4	$5 \cdot 10^{-2}$
^{235}U	$2 \cdot 10^{-4}$	~ 10^{-3}	$3 \cdot 10^{-6}$ (HEU)	~ 10^{-5}	~ 10^{-3}	~ 10^{-5}
^{238}U	$3 \cdot 10^{-2}$	$4 \cdot 10^{-3}$	~ 10^{-7} (DU)	$3 \cdot 10^{-5}$	$3 \cdot 10^{-3}$	
Pu, ^{241}Am	$2 \cdot 10^{-4}$	$5 \cdot 10^{-4}$	$5 \cdot 10^{-6}$	~ 10^{-6}	$2 \cdot 10^{-3}$	

3.2 Radionuclide Anomaly as a Signature and Object under control

- The express analysis is especially used for on-site time-limited diagnostics of characteristics of radionuclide anomalies (RNA) conditioned by the presence of trace (near-background) and weight quantities of radionuclides.
- The control study involves the detection (search and localization) of RNA in a given area and its identification through determining isotope composition and activity of sources.
- The ratio of concentrations of indicator radionuclide typical background in the given environmental object to its detection threshold can be taken as a generalized estimate of detection efficiency ($k_{det.}$).

$$k_{det.} = c_{bg.} / MDC,$$

$c_{bg.}$ is the background concentration at the control point;
MDC is the minimum detectible concentration of radionuclides inducing RNA.

- Value $k_{det.} \geq 1$ indicates that detection system sensitivity is sufficient for recording near-background radionuclide concentrations.

Table 5 – Estimates of relative coefficients, $k_{det.}$, of radionuclide detection in environmental objects.

Radionuclides	Atmospheric Air	Waters	Soil, sediments
^{3}H	1…10	$10^{3}…3\cdot10^{3}$	–
^{90}Sr	0,01…30	$5\cdot10^{3}…10^{6}$	50
^{137}Cs	$3\cdot10^{-3}$ …0,1 (0,3*)	10… $2\cdot10^{3}$	300
^{235}U	0,01…1,0	$10^{2}…5\cdot10^{3}$	1,3…2
$^{234, 238}U$	≈ 30	$\approx 2\cdot10^{5}$	30…50
Pu,	$5\cdot10^{-3}$ …0,1	$\approx 3\cdot10^{3}$	5
^{241}Am	10^{-4}…0,03	$\approx 10^{2}$	1,5

3.3 Detection systems

Nuclides in radioactive materials are the sources of particulate (α, β, n, fission fragments) and quantum (γ, X-ray, bremsstrahlung) radiations. All these radiations (basically, apart from X-ray and especially bremsstrahlung ones) refer to nuclear radiations since their origin is associated to the transformations in nuclei of substances.

When detecting/identifying RNA, means of radiation detection involve small-sized (including portable) instruments, onboard modules used in ground and water carriers, and stand-alone instruments.

They are configured according to the following schematic (Fig. 1).

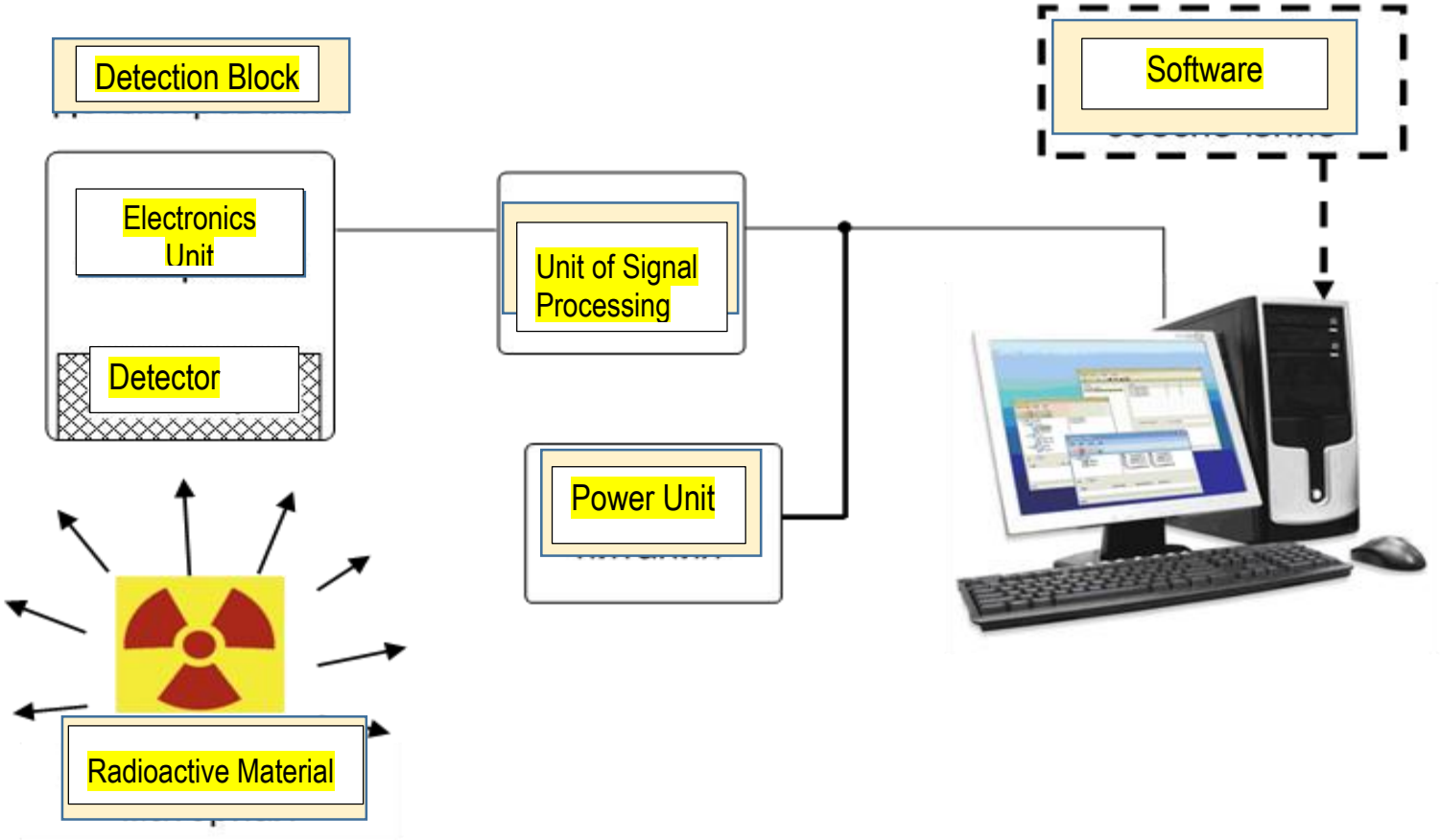
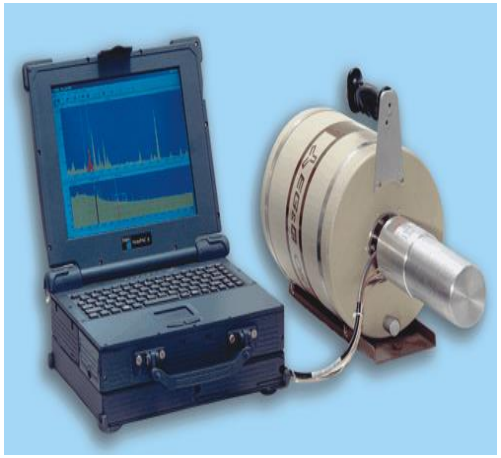
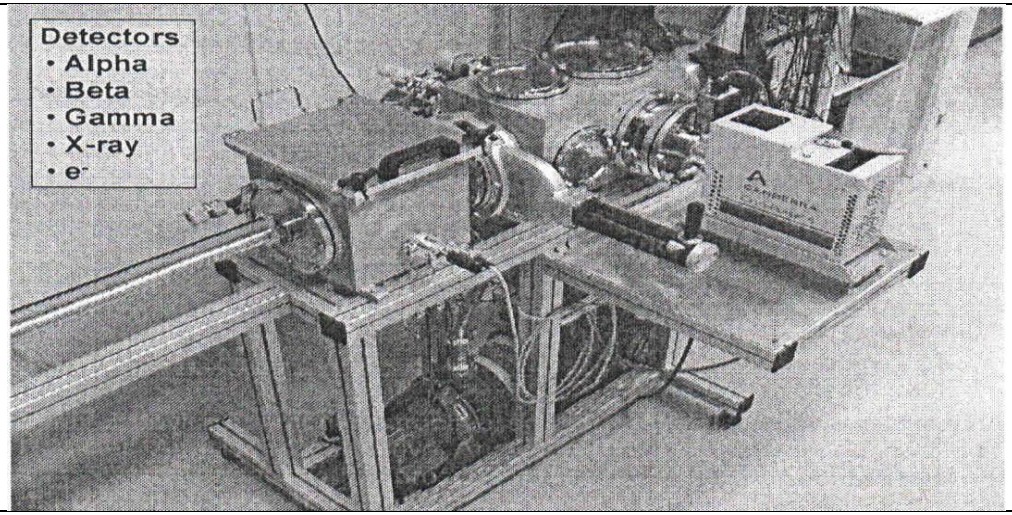


Fig. 1 – Block-diagram of nuclear radiation registration.

Scintillation (NaI(Tl))
spectrometer «Gamma -1C/NB»



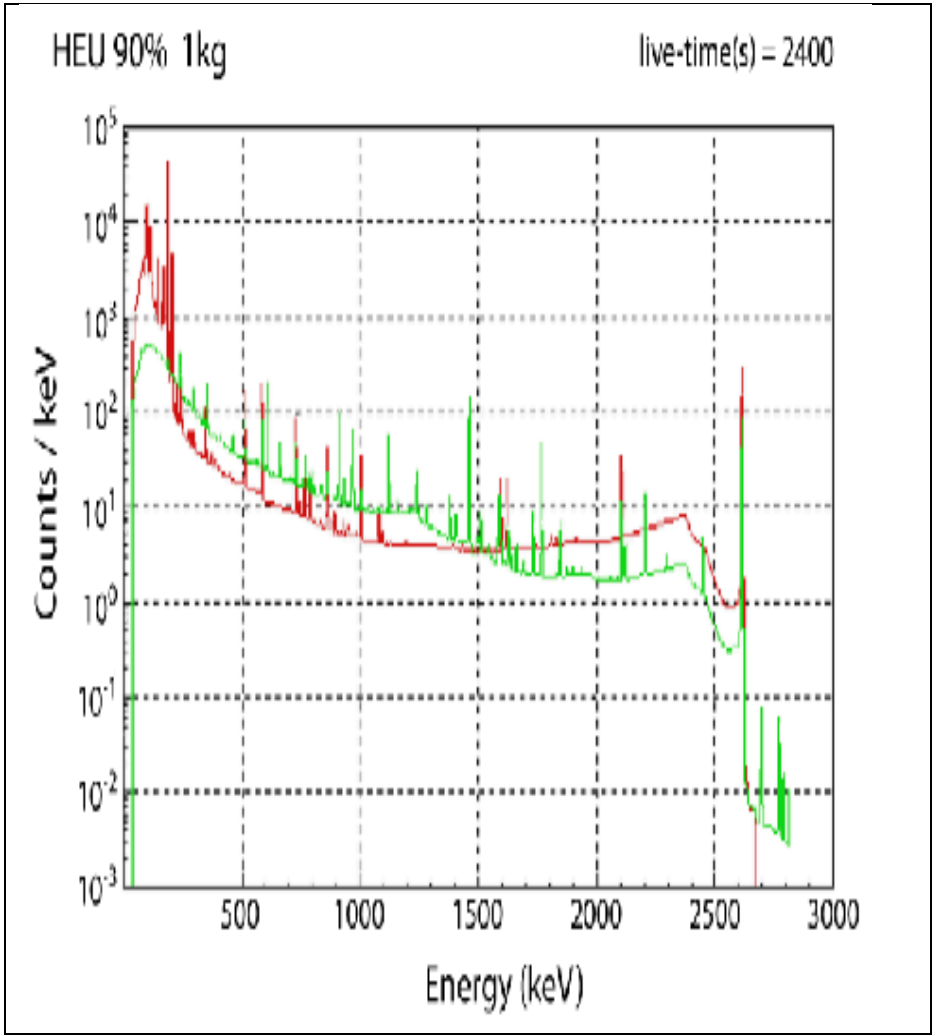
Semiconductor (HPGe) spectrometers (Canberra, ORTEC)



Alpha, beta, gamma, X-ray – emissions
detector module PANDA

Fig.2 – Blocs and modules of radiation detectors.

3.4 Remote gamma-spectrometry (non-destructive analysis) of special nuclear materials using semiconductor HPGe detectors.



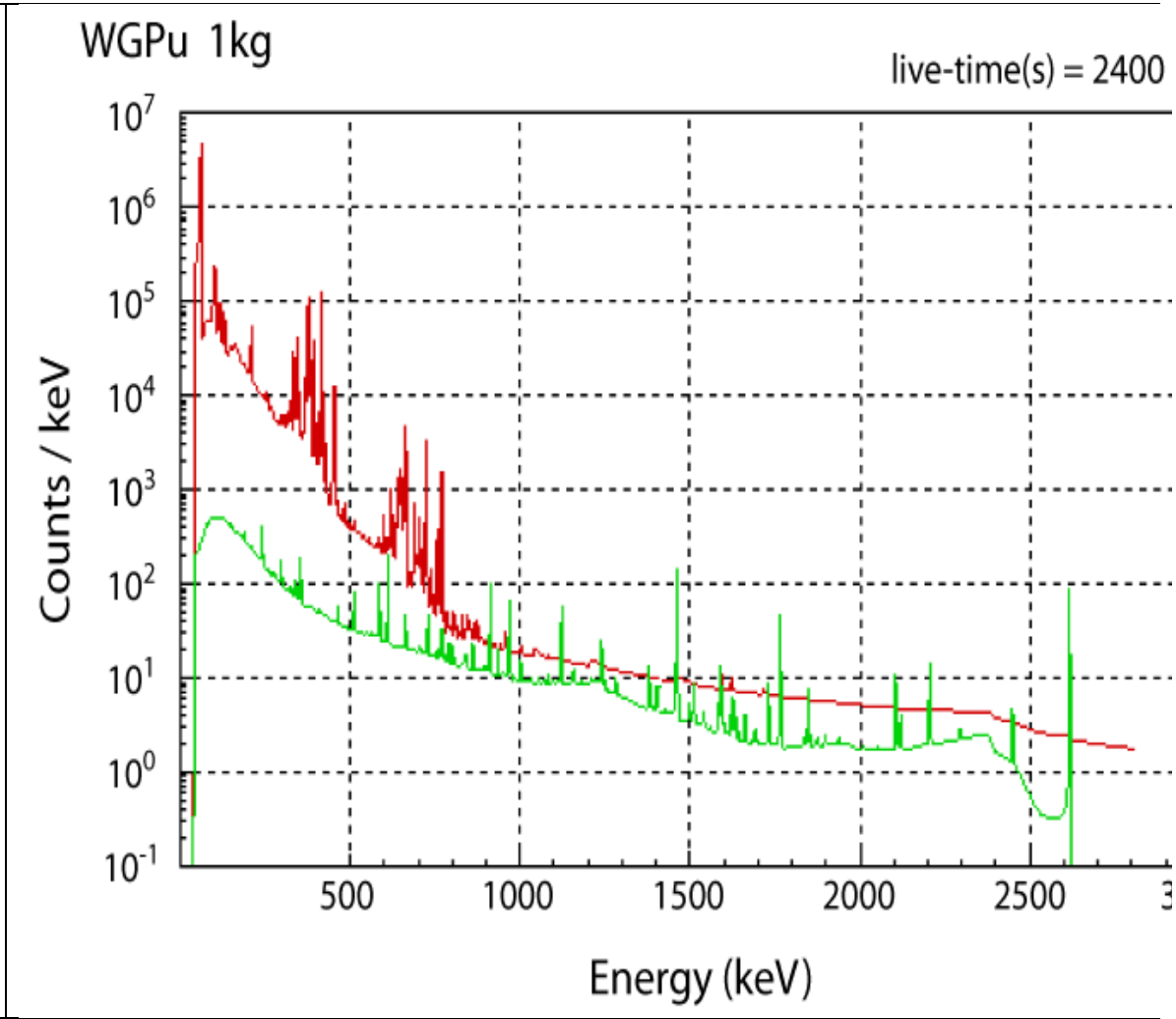


Fig. 3. Gamma-spectra of WGU (left), WGPu (right) (1m distance) and detector background*.

**/J. Medalia. Detection of Nuclear Weapons and Materials: Science, Techologies, Observations. Congressional Research Service, Nov.2008, pp.4-5 /.*

3. Examples of practical implementation

4.1 Investigation into implosion in *RaLa experiments, Los-Alamos (in years 1944-1962)*

In 1944, E.I. Zababakhin, after he had graduated from the academy, continued his education in graduate military course. His Thesis (defended in 1947) was entitled "Investigation into the Processes in Convergent Shock-Waves". Evgeny Ivanovich was one of the most reputable specialists in explosive gas-dynamics, he took active part in development and testing of Soviet atomic bomb.

The first use of radionuclide analysis for implosion study: RaLa experiments to measure uniformity of generated shock-wave (Bayo canion in Los-Alamos) during ^{240}Pu-related crisis.* Idea: R. Cerber (1943), director: B. Rossi, participant: T. Hall (USSR agent).** Gamma-radiation of ^{140}La samples subjected to implosion was recorded (1.8 mg; 1000 Ci / 7TBq). Proportional counters were used (8 pieces; 5 cm in diameter, 76 cm in length, Ar+CO_2 4.5 atm.); liquid scintillators were used later.

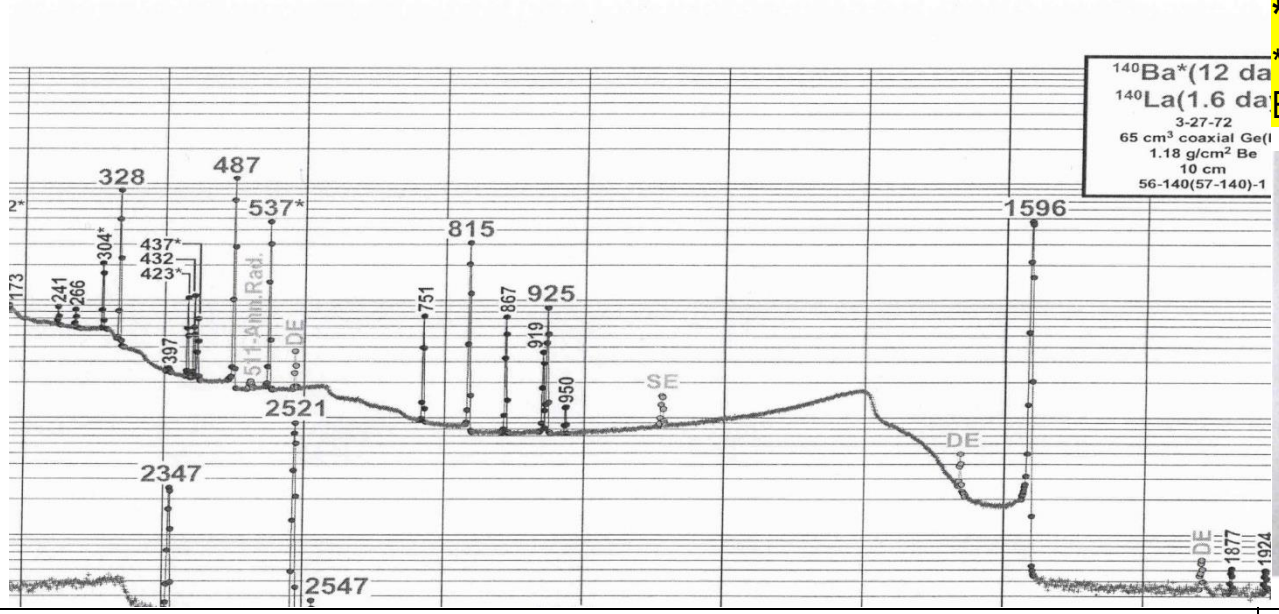


Fig.4. Gamma spectrum ^{140}La (+^{140}Ba).

*http://military.wikia.com/wiki/RaLa_Experiment.

** Richard Rhodes. THE MAKING OF THE ATOMIC BOMB *(pp.312-315). M: Эксмо, 2011.*

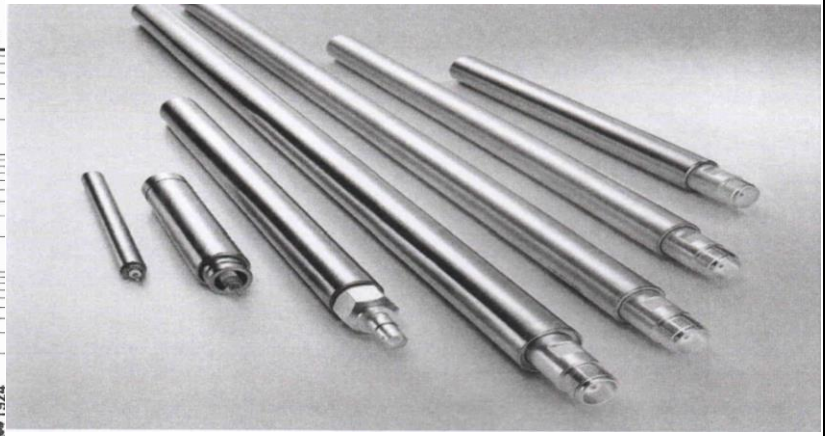
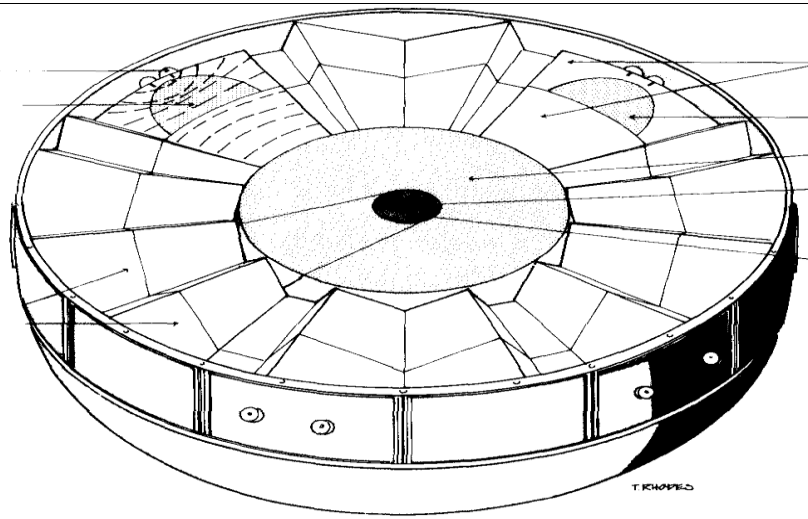


Fig.5. Proportional counters.

Detonator
Detonation valve

High-explosive blocs



«Composition B (TATB) (fast)»
«Baratol» (slow)
Natural uranium tamper
INITIATOR (Po-Be)
^{239}Pu Core (6 kg)

Fig.6. Scheme of earliest implosion-type nuclear explosives* **(Gadget/Trinity, Fat Men; РДС-1).**
*****(D. Rhodes. The making of the Atomic Bomb. A TOUCHSTONE BOOK, 1986. p.575)

Fig.7. Implosion system and registration blocs.

Fig.8. Remote handling registration blocs of ^{140}*La gamma-radiation.*

4.2 Gamma-spectrometry for analysis of radiations from nuclear warhead and slag sample from UNE.

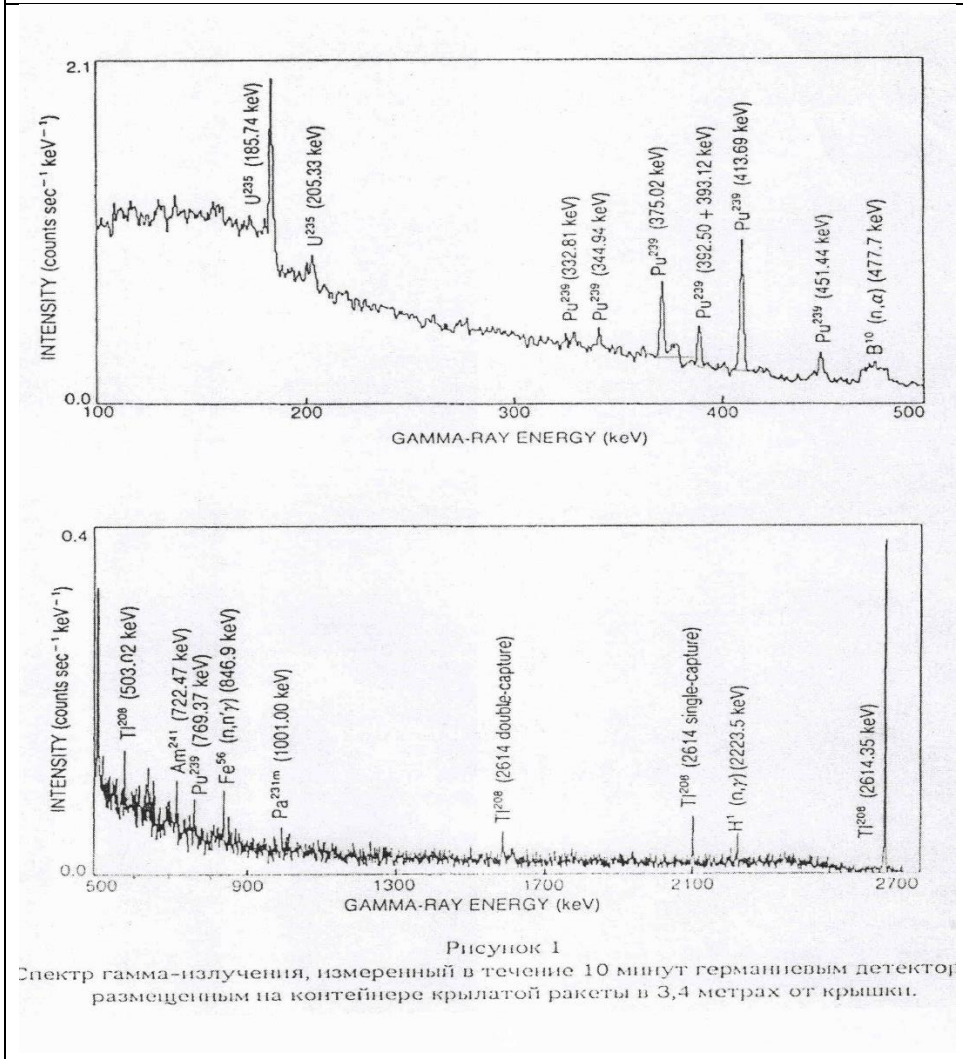


Рисунок 1

Спектр гамма-излучения, измеренный в течение 10 минут германиевым детектор размещенным на контейнере крылатой ракеты в 3,4 метрах от крышки.

Fig.9. Gamma-spectrum of radiation from USSR cruise missile on the deck of "Slava" cruiser (1989).*

**/Fetter C, Fon Hippel F. Science and Global Security. 1990, V.1, No`s 3-4, p.p.323-333/.*

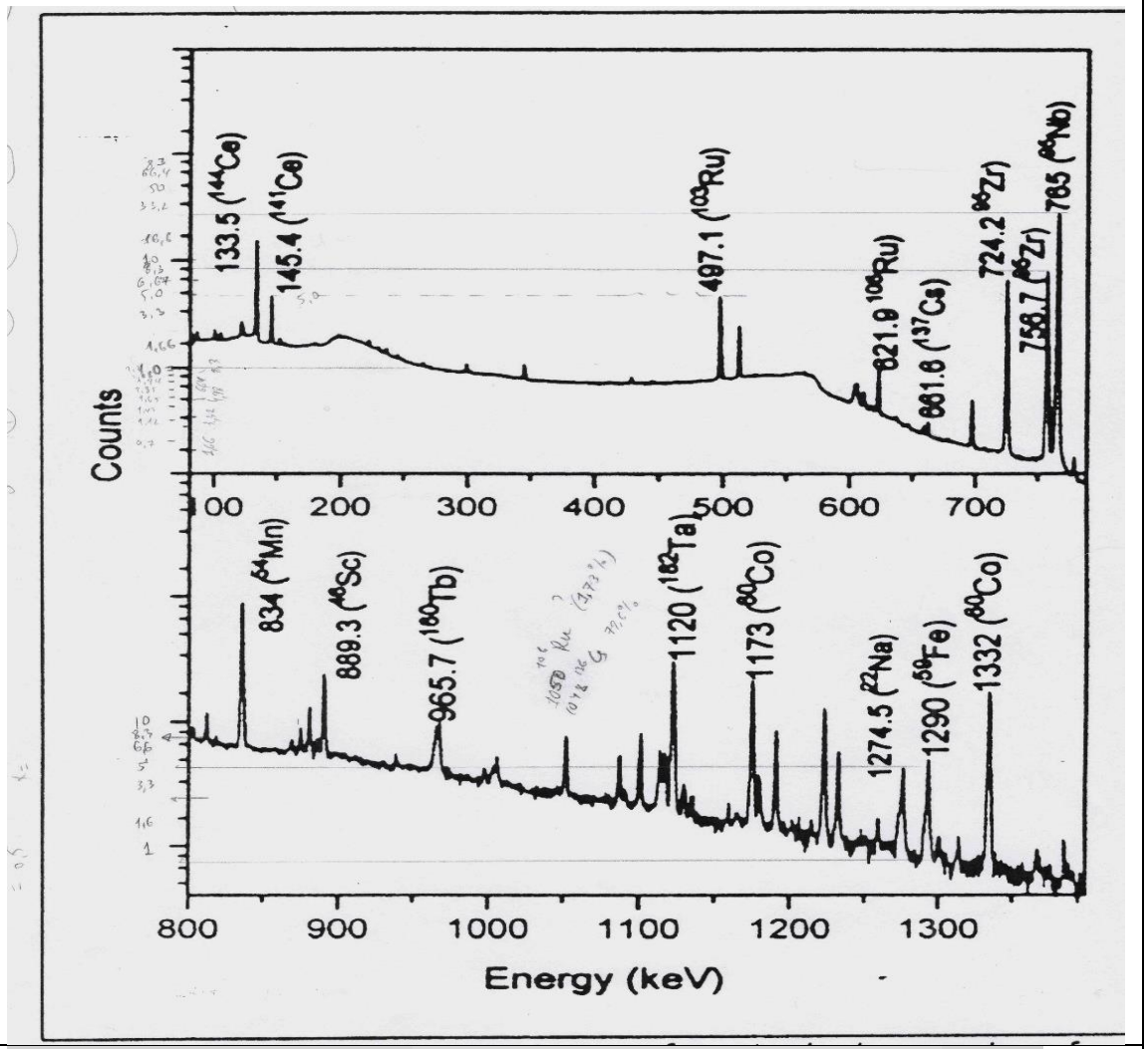


Fig.10. Gamma-spectrum of slag sample from UNE in India, 1998.*

**/S.B Manhar at al. BARC NEWSLETTER. No.186, July 1999.pp. 1-6/*

4.3 Direct X-ray and alpha-spectrometry of test ^{239}Pu + ^{241}Am and “thick” DU samples.

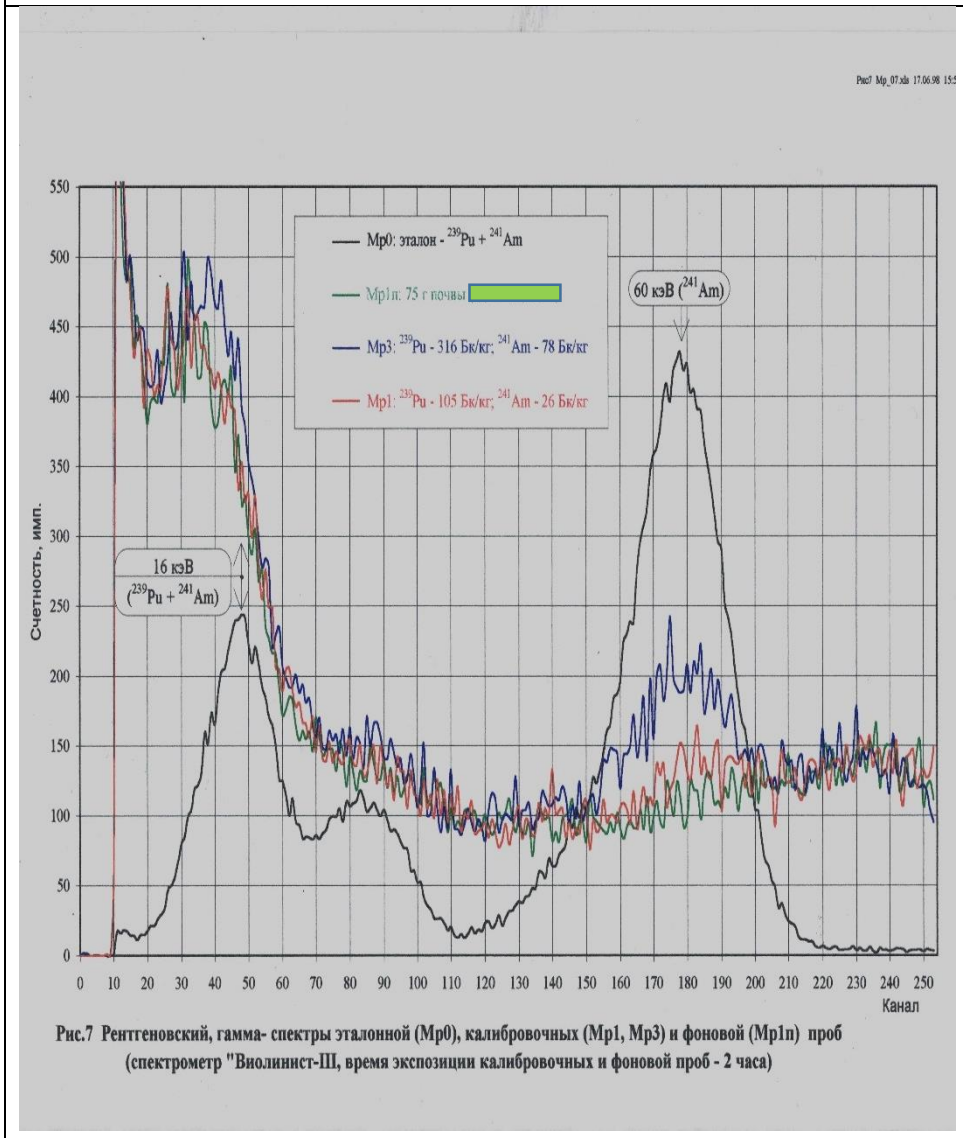


Рис.7 Рентгеновский, гамма- спектры эталонной (Mp0), калибровочных (Mp1, Mp3) и фоновой (Mp1n) проб (спектрометр "Виолинист-III, время экспозиции калибровочных и фоновой проб - 2 часа)

Fig. 11 . Gamma- and X-ray radiation spectra of friable soil (sand) samples containing ^{239}Pu+^{241}Am mixture.

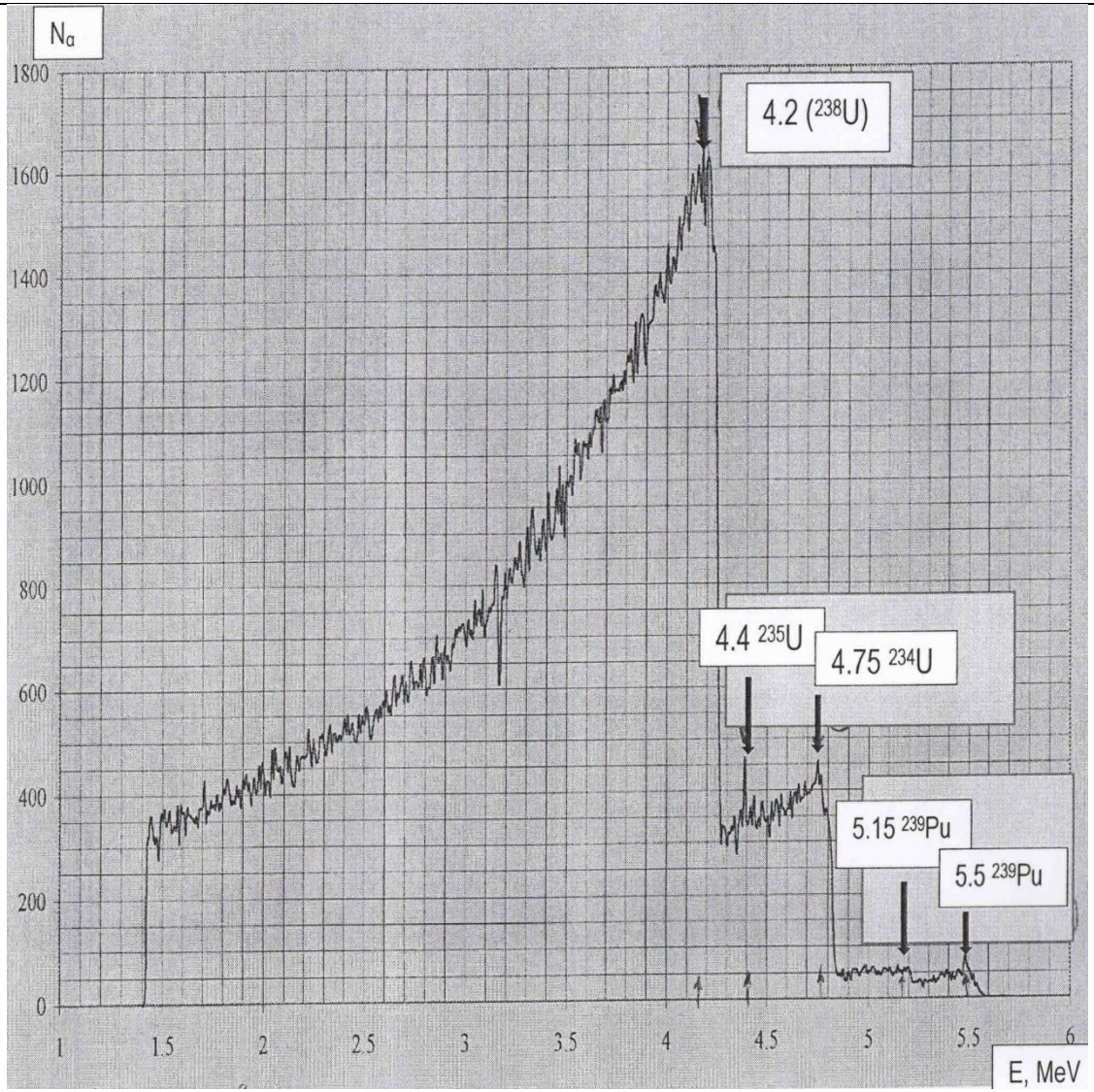


Fig.12. Alpha-spectrum of “thick” layer DU sample with trace quantities of $^{234,235}U$ and ^{238}Pu.

4.4 Gamma-spectra of radionuclides in environmental soils using scintillator and semiconductor detectors.

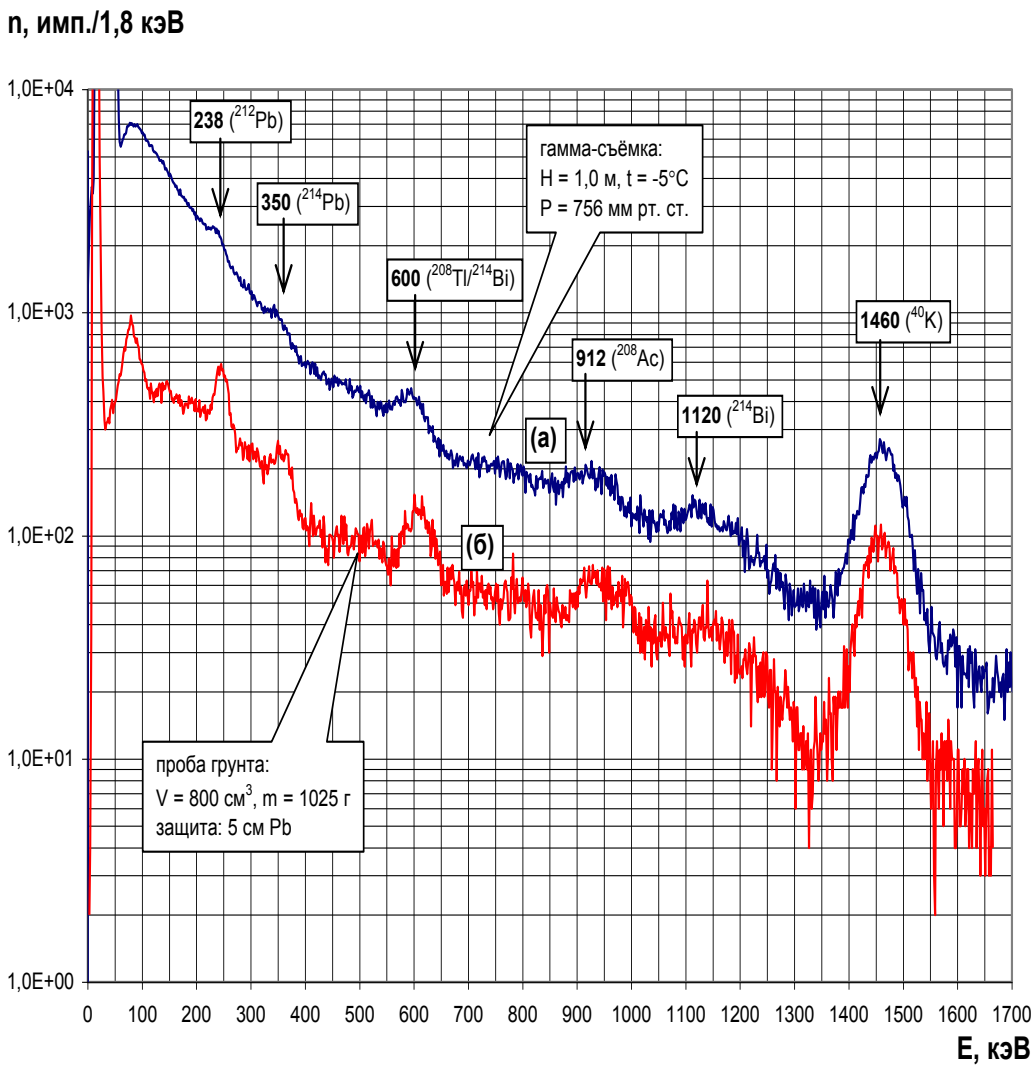


Fig.13. Gamma-spectra of surface survey (a) and soil sample (b). Detector NaI(Tl) Ø63x63 cm³ (expose time - 1 hour)

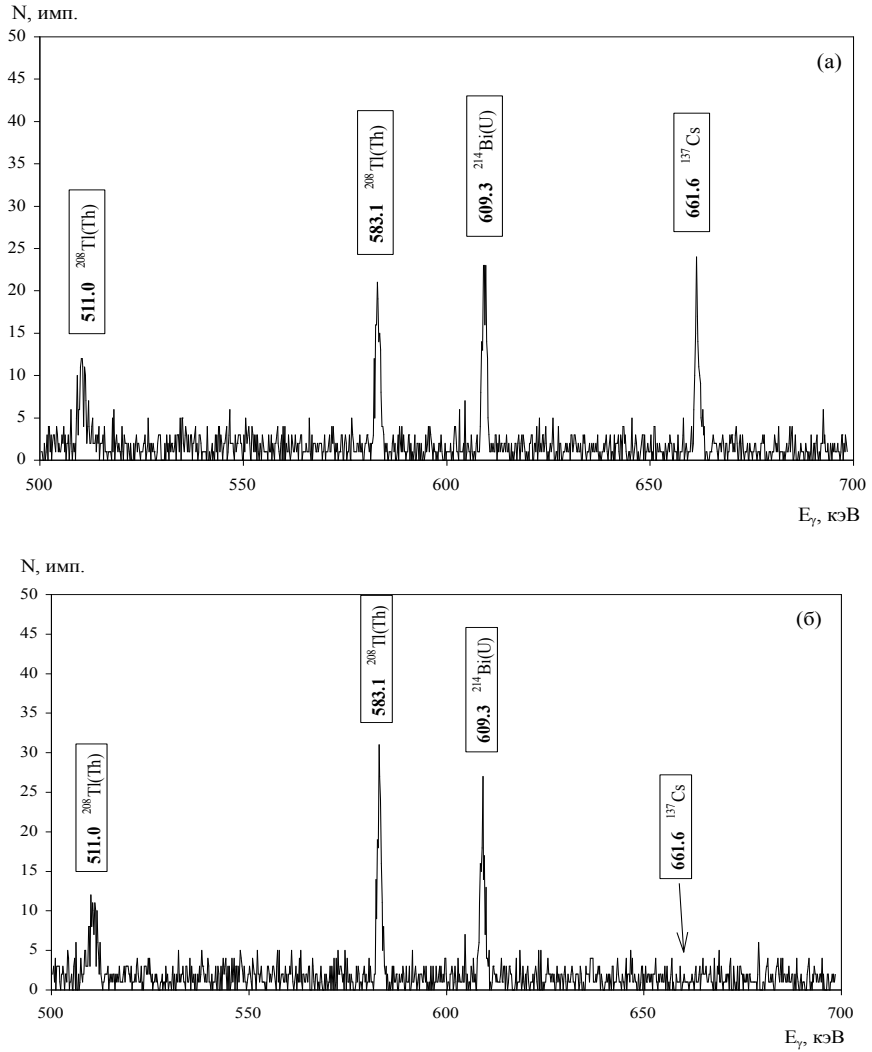


Fig.14. Gamma-spectra «windows» of soil samples from surface (top) and from 5 m depth (below). Detector - HPGe.

4.5 Detection of ^{90}Sr specific content in soil samples using direct instrumentation method (DIM)

DIM is based on the measurements of summing β-activity of soil preparation samples and γ-spectrometric determination of specific content of ^{137}Cs and natural radionuclides in this samples.

DIM threshold sensitivity is estimated to be equal to $a_{\beta,min}(^{90}Sr) \approx 50$ Bq/kg which corresponds to the soil contamination value of A_s (^{90}Sr)≈0,3 Ci/km^2 (10 кBq/m^2).

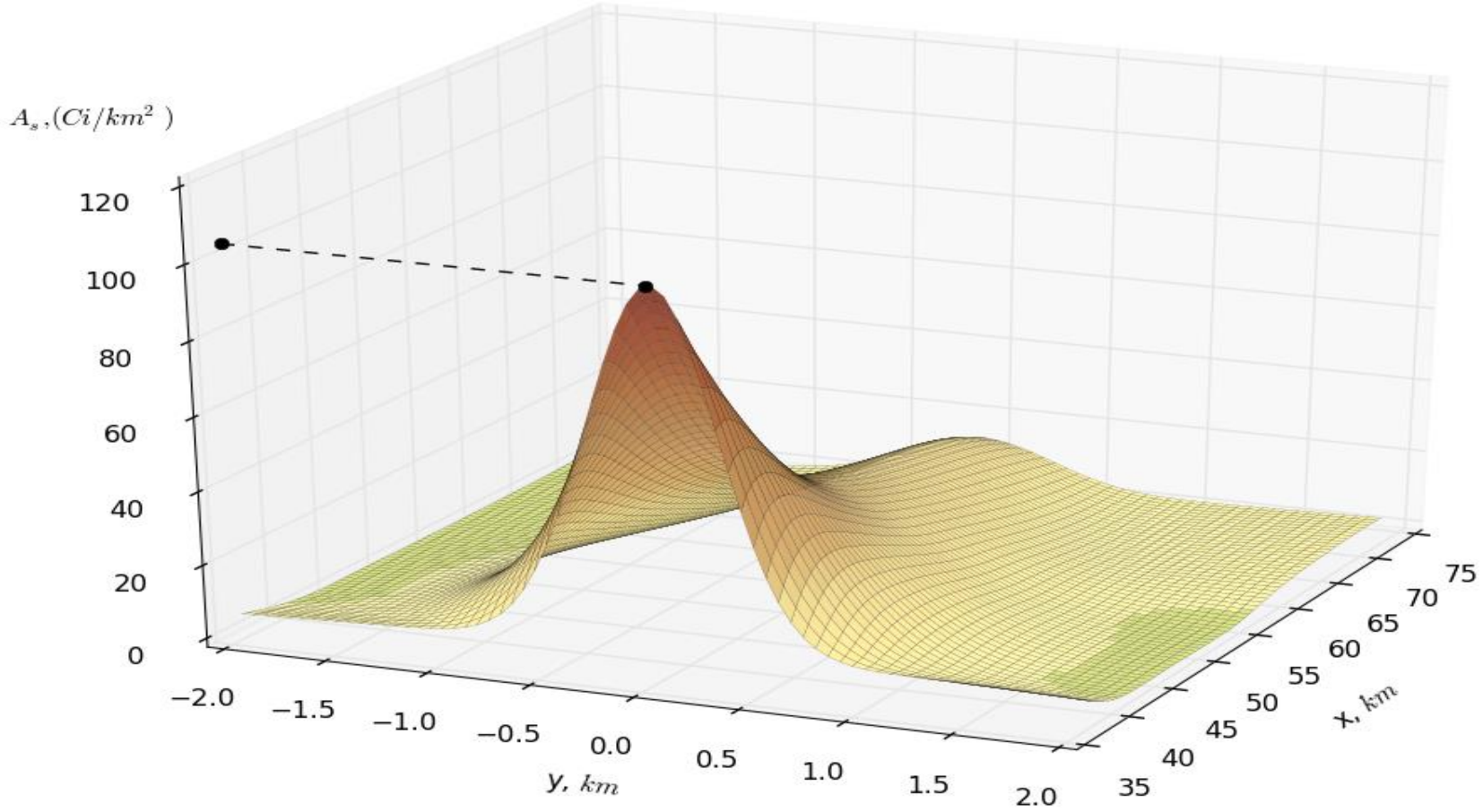


Fig. 15. Model relief of ^{90}Sr contaminations in the middle part of Eastern-Ural Radioactive Trace (at the accident explosion time)*.
**/В.В. Власов, Н.В. Ивашкин, А.Л. Карманов. Препринт РФЯЦ-ВНИИТФ №254, 2016 г./*

4. Conclusions

Radionuclide express analysis is one of the key components of the technical system used to control nuclear materials and high-activity sources in civilian and weapons cycles and intended to provide local and global safety and security.

This analysis should be preferably used as means of on-site inspection of numerous objects.

The express analysis in some cases exhibits a worse detection sensitivity if compare to the methods and technical means of stationary analytical laboratories but, being used on-site with stand-alone modules, it shows a sufficient level of selectivity and proves to be cost- and time-effective in large-scale measurements.

The most conclusive methods i.e. remote gamma-spectrometry and non-destructive in-situ analysis samples and objects can be modified in control systems depending on non-intrusive requirements to meet bilateral, multilateral, and international agreements.