GAMMA-SPECTROMETRY IN IDENTIFICATION OF ARCHAEOLOGICAL SAMPLES

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The abilities of semiconductor γ-spectrometry is solving some problems of archaeology, in particular, the geography of economical relations between different regions of Transcarpathia and adjacent areas during 5—6 millennia before Christ have been considered. Some models for the development of γ-spectrometric taxonomic patterns are discussed. These models are based on: the ratios of relative specific activities (RSA) of γ-active nuclides (GAN) included into the Th-232, U-238, U-235, Np-237 series; the RSA ratios for GAN-representatives of different natural series (e. g. U-238 — GAN / U-235 — GAN); the RSA ratios not included in to the Th- and U-, Np-series (K-40, Cs-137, Na-22, Co-60 etc.).

The analysis is done on the basis of experiment carried out at the 36 samples of pottery (100 cm^3 Ge(Li)-detector, passive (Pb-Cu-Al-Cd-Fe) protection, 4 hour exposure). The methodological questions, in particular, the correct account for the experimental background — have been considered on the basis of the analysis of the post-Chornobyl GAN (Cs-137, Ag-110m etc.) content.

Gamma-activity of background (BGA) is a component of accurate measurements in applied nuclear gamma-spectrometry (ANGS). At the experimental complex of the Photonuclear Process Department of the Institute of Electron Physics. Ukr. Nat. Acad. Sci., where the electron accelerator microtron M-30 is operated, BGA is determinated by the material of the biological protection and is registrated by the gamma-spectrometry equipment. In general, the integral BGA may be presented by the dependence:

$\sum_i (1 - e^{\lambda_i T_I})$, where T_i is the duration of M-30 operation.

$\sum_i e^{\lambda_i T_D}$, where T_D is the duration of technological pauses of M-30 operation.

$\sum_i (1 - e^{\lambda_i T_C})$, where: T_C is the duration of BGA measurements; i is the amount of gamma-active nuclides (GAN); $\lambda_i = \ln 2 / T_{1/2}$ is the decay constant with $T_{1/2}$ = seconds ÷ years.

In general, the BGA spectral composition has been interpreted with such GAN-lines (E_γ, keV):

Th^{234}: 63, 93; Pb^{214}: 295, 352; Bi^{214}: 609, 112, 1764 (1);

these GANs are the GANs of U^{238}-series (underline means stable nuclide):

$_{92}U^{238}$, $_{92}U^{234}$; $_{91}Pa^{234}$, $_{91}Pa^{234m}$; $_{90}Th^{234}$, $_{90}Th^{230}$; $_{88}Ra226$; $_{86}Rn^{222}$, $_{86}Rn^{218}$; $_{84}Po^{210}$, $_{84}Po^{214}$, $_{84}Po^{218}$; ; $_{84}Po^{210}$, $_{84}Po^{214}$, $_{84}Po^{218}$; $_{83}Bi^{210}$, $_{83}Bi^{214}$, $_{83}Bi^{210m}$; $_{82}\underline{Pb}^{206}$, $_{82}Pb^{210}$, $_{82}Pb^{214}$; $_{81}Tl^{206}$ (2);

with lines: U^{235}: 143.8; 185.7 (3);

these GANs are the GANs of U^{235}-series:

$_{92}U^{235}$; $_{91}Pa^{231}$; $_{90}Th^{231}$, $_{90}Th^{227}$; $_{89}Ac^{227}$; $_{88}Ra^{223}$; $_{87}Fr^{213}$; $_{86}Rn^{219}$; $_{85}At^{215}$; $_{84}Po^{215}$, $_{84}Po^{211}$; $_{83}Bi^{211}$; $_{82}Pb^{211}$, $_{82}\underline{Pb}^{207}$; $_{81}Tl^{207m}$, $_{81}Tl^{207}$ (4);

with lines: Tl^{208}: 277, 328, 338, 583, 2614; Pb^{212}: 239; Ac^{228}: 209, 270, 911, 969 (5);

these GANs are the GANs of Th^{232}-series:

$_{90}Th^{232}$, $_{90}Th^{228}$; $_{89}Ac^{228}$; $_{88}Ra^{224}$; $_{86}Rn^{220}$; $_{85}At^{216}$; $_{84}Po^{216}$; $_{83}Bi^{212}$; $_{82}Pb^{212}$, $_{82}Pb^{208}$; $_{81}Tl^{208}$ (6),

and also by GAN-lines which are the gamma-active fission products (GAFP): Cs^{137}: 662 (7),

and also Co^{60}: 1173, 1332; K^{40}: 1461 (8).

In case of activation analysis (AA) it is necessary also to measure BGA of samples (BGA_S): BGA_{S1} — before the irradiation of samples: BGA_{S2} — after AA. If there are appreciable U and Th quantities in samples then $BGA_{S2} > BGA_{S1}$.

In general, BGA_S has been also interpreted by the lists (1, 3, 5, 7, 8). So, if for determination of specific activities (SA) of BGA the statistical criteria of photopeak selection (and identification)

$$\alpha_B = \frac{\Delta S_B}{\sqrt{\Delta S_B + 2F}} \geq 1; \qquad (9)$$

was used; then, for determination of SA of BGA_S the statistical criteria of photopeak selection (and identification)

$$\alpha_S = \frac{\Delta S_S}{\sqrt{\Delta S_S + 2\Delta S_B}} \geq 1, \qquad (10)$$

was used, where ΔS_B is a photopeak area in the case of BGA measurements; ΔS_S is a photopeak area in the case of BGA_S measurements; F is the area of the base under the photopeaks.

The composition of BGA and BGA_S was essentially changed after Chornobyl accidents (April—August 1986; 1991; 1994; 1995), when the appreciable GAFP-quantity in the environment were registered:

Zr^{95}; Nb^{95}; Ru^{103}, Ru^{106}; I^{129}, I^{131}, I^{132}, I^{133}, I^{135}; Cs^{134}, Cs^{137}; Ce^{141}, Ce^{144}; Eu^{152}, Eu^{154}. (11)

A big part of GAFP in Transcarpathia was stopped in the Carpathian mountains and forests. The Chornobyl accidents made the lists (1, 3, 5, 7, 8) to be corrected with the consideration of the list (11). The following list of GANs is used from 1992 (E_γ, keV, GAN):

186.1 — Ra^{226}; 276,4 — Ba^{133}; 295.1 — Pb^{214}; 427.95 — Sb^{125}; 583.1 — Tl^{208}; 609.3 — Bi^{214}; 662 — Cs^{137}; 795.8 — Cs^{134}; 884 — Ag^{110m}; 911 — Ac^{228}; 1063.6 — Bi^{207}; 1173.2 — Co^{60}; 1274.5 — Eu^{154}; 1460.75 — K^{40}; 1620.8 — Bi^{212}. (12)

Specifically, list (12) was used for BGA_S determination in human blood [1]. It was made the passive combined (Pb-Cu-Al-Cd-Fe) protection with the aim to make prolonged stability (within the 5% limit is) and decreasing of BGA. After experimental researches it becomes possible to work with $\Delta S_S \geq 10$ ($\alpha_S \leq 1$) and to solve difficult analytical problems.

It is possible to understand the list (12) as the experimental pattern of samples, though there are some analytical problems for which this list is not full. Specifically, these are:

1. The problems of the "black box" samples recognizing (illegal transfer of radioactive materials). The list (12) has been expanded by GAN of Pu, Am, Cm, Cf and their GAFP.

2. The problems of industrial transfer of radioactive nuclides (RAN) (connected with industrial processes) [2].

3. The problems of soil erosion and landslips in Transcarpathia. Therefore it is necessary to measure the following GAN: U^{232}, U^{235}, U^{236}, U^{238}; Pu^{236}, Pu^{238}, Pu^{239}, Pu^{240}, Pu^{242}; Np^{237}; Am^{241}, Am^{242m}, Am^{243}; Cm^{242}, Cm^{243}, Cm^{244}, Cm^{245}, Cm^{246} and their GAFP.

4. The problems of monitoring of tectonic activity. To solve this problem it is necessary to measure "low-" and "high-" frequent BGA variations (specifically, U^{234}, Rn^{218}, Rn^{222}).

5. The problems of archaeology, in particular, the geography of economical relations between different regions of Transcarpathia and adjacent areas during 5—6 millennia before Christ. The elements of argil pottery (36 samples, 100 cm^3 Ge(Li)-detector, passive Pb-Cd-Cu-Al protection, 4 hour exposure) were used as the samples in this case.

This problem is difficult because the time, the place of potters manufacture and their using are unknown; these samples were in the ground at a depth of 0.5—1.5 m. To solve these problems it is necessary to select the models which could be used as the experimental pattern of samples. They are:

1. Determination of relative specific activities (RSA) of the GANs of U-, Th-series:

U^{238}: Pb^{214}, Bi^{214}, Ra^{226};
Th^{232}: Pb^{214}, Ta^{208}, Ac^{228}.

The RSA ratio of GAN for each of series will make it possible to estimate the radioactive equilibrium (for each of series).

2. It is possible to show the U-, Th-series as:

$$U^{238} \rightarrow \underline{Pb}^{206} + 8\ He^4;$$
$$U^{235} \rightarrow \underline{Pb}^{207} + 7\ He^4;$$
$$Th^{232} \rightarrow \underline{Pb}^{208} + 6\ He^4.$$

It possible to estimate:
the radiogenic stable Pb^{206} quantity using RSA of Bi^{210}, Pb^{210}, Tl^{206}, Tl^{210};
the radiogenic stable Pb^{207} quantity using RSA of Bi^{211}, Pb^{211}, Tl^{207};
the radiogenic stable Pb^{208} quantity using RSA of Bi^{212}, Pb^{212}, Tl^{208}, Po^{212};

because these GANs are located in the end of U-, Th-series. Determination of Pb^{208}/Pb^{206}-, Pb^{207}/Pb^{206}-ratios is used to solve the archaeological problems.

3. Determination of RSA of the GAN: Cs^{137}, Na^{22}, Co^{60}, K^{40}.

To solve the problems of such type it is necessary to expand the list of GAN. Experimental researches of this list will allow to estimate the limit is of using methods. The offering list will be ($T_{1/2}$ > 100 years; in brackets — natural contents, %):

$_{11}Na^{22}$, $_{13}Al^{26}$, $_{19}K^{40}$ (0.0117), $_{23}V^{50}$ (0.250), $_{27}Co^{60}$, $_{36}Kr^{81}$, $_{40}Zr^{96}$ (2.8), $_{41}Nb^{96}$, $_{41}Nb^{92}$, $_{41}Nb^{94}$, $_{42}Mo^{93}$, $_{41}Nb^{93m}$, $_{43}Tc^{97}$, $_{43}Tc^{98}$, $_{47}Ag^{110m}$, $_{50}Sn^{126}$, $_{51}Sb^{126m}$, $_{51}Sb^{126}$, $_{52}Te^{123}$ (0.908), $_{53}I^{129}$, $_{57}La^{137}$, $_{57}La^{138}$ (0.09), $_{67}Ho^{166m}$, $_{71}Lu^{176}$ (2.59), $_{72}Hf^{182}$, $_{73}Ta^{182}$, $_{73}Ta^{180}$ (0.012), $_{74}W^{180}$ (0.13), $_{75}Re^{186m}$, $_{75}Re^{186}$, $_{77}Ir^{192m2}$, $_{77}Ir^{192}$, $_{80}Hg^{194}$, $_{79}Au^{194}$. (13)

It is necessary also to research GAN of Np-series:

$_{93}Np^{237}$; $_{92}U^{233}$; $_{91}Pa^{233}$; $_{90}Th^{229}$; $_{89}Ac^{225}$; $_{88}Ra^{225}$; $_{87}Fr^{221}$; $_{85}At^{217}$; $_{83}Bi^{213}$; $_{81}Tl^{209}$ ($?_{83}\underline{Bi}^{209}$). (14)

Table 1. Relative specific activities RSA for samples NN 4, 8, 9, 10, 19 (average energy E , 50—200 keV)

E, keV	RSA, r.u.					
	N4	N8	N9	N10	N19	Possible GANs
55	B	—	1.96	5.07	B	Re^{186m}, At^{218}, Fr^{223}, Th^{227}, Th^{232}, U^{234}, Am^{241}
67	13.88	24.3	16.7	9.7	15.8	Ac^{225}, Th^{230}, Th^{234}, Pa^{234}
80	—	B	23.71	15.15	B	Ba^{133}, Eu^{155}, Ho^{166m}, Fr^{223}, Th^{227}, Th^{228}
90	33.52	B	20.87	12.21	B	Lu^{176}, Ta^{180}, Th^{227}, Th^{234}, Np^{237}
100	—	2.05	B	0.45	1.9	Ta^{182}, Re^{186m}, Fr^{221}, Fr^{223}, Ac^{222}, Th^{231}, Pa^{234}
109	B	B	B	—	1.7	Eu^{155}, U^{235}
118	4.96	—	—	—	4.85	Ta^{209}, At^{216}, Fr^{221}, Np^{237}
122	—	—	—	—	2.08	Eu^{152}, Eu^{154}, Re^{186}, Ra^{223}, Th^{229}, Th^{232}, U^{234}
132	8.56	3.23	—	B	—	Ce^{144}, Re^{186}, Rn^{219}, Fr^{221}, Th^{228}, Th^{229}, Pa^{234}
144	B	B	B	B	B	Ra^{223}, Ac^{225}, Th^{229}, Th^{230}, U^{235}, Np^{237}
152	—	5.59	—	1.47	5.56	Hf^{182}, Ta^{182}, Ta^{223}, Ac^{225}, Th^{229}, Pa^{224}, Np^{237}
163	3.74	4.96	—	0.94	2.71	Th^{231}, Ir^{192m}, Ac^{227}, U^{235}
177	B	B	B	B	2.74	?
186	7.05	1.26	2.47	1.65	5.03	Ra^{226}, Ho^{166m}, Ac^{225}, U^{235}
195	B	0.55	—	2.07	—	Th^{229}, U^{235}, Np^{237}

B: BGA = BGAs; —: photopeak is not selected; ?: photopeak is not identified.

References

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ГАММА-СПЕКТРОМЕТРІЯ ДЛЯ ІДЕНТИФІКАЦІЇ АРХЕОЛОГІЧНИХ ЗРАЗКІВ

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Розглянуто можливості напівпровідникової гамма-спектрометрії для розв'язання деяких питань археології, зокрема географії господарських зв'язків між різними регіонами Закарпаття та прилеглих регіонів у період 5–6 тисячоліття до н. е. Розглядаються деякі можливі моделі для створення гамма-таксонометричних образів, зокрема, на основі співвідношень відносних питомих активностей (ВПА) гамма-активних нуклідів (ГАН),що входять в один з рядів Th^{232}, U^{235}, U^{238}, Np^{237}; на основі співвідношень ВПА ГАН – представників різних рядів (напр., U^{238} і U^{235}); на основі співвідношень між ГАН, які не входять до названих рядів (K^{40}, Cs^{137}, Na^{22}, Co^{60} і т.д.). Розгляд виконується на основі експерименту, виконаного на 36 зразках гончарних виробів (100 $см^3$, ДГДК-детектор; пасивний (Pb-Fe-Cd-Cu-Al) захист; тривалість одного виміру 4 год). Розглянуто методичні питання, зокрема, коректне врахування фону експериментального обладнання на основі аналізу вмісту "постчорнобильских" ГАН (Cs^{134}, Ag^{110m} та ін.).