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GAMMA SPECTROMETRY IN THERMOLUMINESCENCE DATING

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Abstract. The aim of this paper is to present in detail a method, adopted in our laboratory, for the determination of annual dose. In particular, an analysis is focused on the thermoluminescence dating of eolian sediments from Kępa Kujawska. The advantage of the presented method is that partial matrix doses for alpha, beta and gamma radiation are determined simultaneously from the high resolution gamma spectrometry measurements.

1. INTRODUCTION

Thermoluminescence (TL) in semiconductors is the luminescence emission stimulated by heating, following the previous absorption of energy from ionised radiation. Many minerals show the so-called natural TL since they have been exposed to the natural nuclear radiation. The duration of the natural radiation exposure, t, i.e. the „age" of an object, can be derived from the general equation

$$t = D / D', \qquad (1)$$

where D' is the dose rate of the radiation and D denotes the total dose accumulated by TL phenomenon (archaeodose or paleodose). In general, the evaluation of paleodose is possible when the relationship between TL energy and absorbed radiation dose from an artificial source is established (Aitken, 1985; Mejdahl and Wintle, 1984).

In natural environment, the major sources of radiation originate from naturally occurring radioactive decay chains and elements: ^{232}Th, ^{238}U, ^{235}U series, ^{40}K, ^{87}Rb isotopes and from cosmic radiation. In evaluation of the rate at which ionisation energy is deposited in a large sample of soil the "infinite matrix" assumption is applied and consequently, the absorption of energy per unit mass is equal to the emission of energy per unit mass.

However, in the determination of the dose rate for TL dating it is important to distinguish between the dose absorbed by the soil matrix and by the grain showing TL (Zimmerman, 1972, Murray *et al.,* 1978; Mejdahl, 1979). For example, quartz grains separated from the soil are almost free of internal radioactivity. The outside layer of large grains, which has absorbed alpha particles, can be etched. The remaining cores, excited by the beta and gamma contributions from the matrix, are used in the quartz inclusion technique of TL dating. On the contrary, the fine-grain technique is concerned only with the grains below 10 μm which have received the total matrix dose.

In particular, water is also the constituent of soil but in comparison with minerals its absorption coefficient per unit mass is higher by 50% for alpha, by 25% for beta particles and by 14% for gamma radiation (Aitken, 1985). Moreover, the TL sensitivity depends on the value of linear energy transfer (LET) and decreases substantially for the alpha component of natural radiation. Therefore, the annual dose rate of radiation has to be divided at least into three contributions related to the varied absorption of alpha, beta and gamma radiation.

In practice of TL dating, the efficiency factors $k(\alpha)$, $k(\beta)$ and $k(\gamma)$ are introduced to describe effect of different radiation on the "effective" dose rate-D'. This results in a modified equation (1) in which the dose rate – D' is determined by

$$D' = k(\alpha)\, D'(\alpha) + k(\beta)\, D'(\beta) + k(\gamma)\, D'(\gamma), \qquad (2)$$

where the coefficients $k(\beta)$ and $k(\gamma)$ are usually close to one, since they describe a weak influence of the grain etching and wetness, but $k(\alpha)$ is about 0.15 (Zimmerman, 1972).

The determination of dose rate from activity measurements requires factors for converting relevant isotope activity to the energy released per disintegration.

For TL dating this energy has to be related to the alpha, beta and gamma contribution of radiation. As usual, Auger and internal conversion electrons are included in the beta part, X-ray and bremsstrahlung radiation in the gamma contribution. Calculation of these factors is based on the values from nuclear data tables (Brown and Firestone, 1986; Negin, 1990). Because of the number of various transitions with different probabilities, the final contribution per disintegration may be the sum of quite large number of components.

The energies given in **Table 1** are the products of the emitted energy and branching factors, then for any chain they correspond to the energy released per parent disintegration (Oczkowski and Przegiętka, 1998).

Table 1. *Branching in the decay chains and energy released per parent disintegration*

Nuclide	Branch.	E_α [keV]	E_β [keV]	E_γ [keV]
^{232}Th	1.000	4005.0	0.0	0.2
^{228}Ra	1.000	0.0	11.6	0.0
^{228}Ac	1.000	0.0	479.0	992.5
^{228}Th	1.000	5399.0	20.1	3.4
^{224}Ra	1.000	5675.0	2.2	10.0
^{220}Rn	1.000	6287.9	0.0	0.0
^{216}Po	1.000	6778.5	0.0	0.0
^{212}Pb	1.000	0.0	175.2	145.0
^{212}Bi	1.000	2174.0	502.5	107.1
^{212}Po	0.641	5630.8	0.0	0.0
^{208}Tl	0.359	0.0	214.6	1211.6
^{238}U	1.00	4194.0	9.5	1.3
^{234}Th	1.00	0.0	15.8	9.4
^{234}Pa(m)	0.9987	0.0	821.6	13.8
^{234}Pa	0.0013	0.0	0.6	2.5
^{234}U	1.00	4773.0	0.0	0.1
^{230}Th	1.00	4665.0	0.0	0.4
^{226}Ra	1.00	4774.0	3.5	6.7
^{222}Rn	1.00	5489.2	0.0	0.0
^{218}Po	1.00	6001.3	0.0	0.0
^{214}Pb	1.00	0.0	293.9	250.2
^{214}Bi	1.00	0.0	662.4	1509.2
^{214}Po	1.00	7686.8	0.0	0.0
^{210}Pb	1.00	0.0	34.2	4.7
^{210}Bi	1.00	0.0	389.0	0.5
^{210}Po	1.00	5304.4	0.0	0.0
^{235}U	1.00	4378.0	42.0	156.0
^{231}Th	1.00	0.0	173.0	29.0
^{231}Pa	1.00	4923.0	48.0	39.9
^{227}Ac	1.00	67.3	12.5	0.2
^{227}Th	0.9862	5820.6	53.3	109.8
^{223}Fr	0.0138	0.0	5.5	0.9
^{223}Ra	1.00	5697.0	73.1	134.8
^{219}Rn	1.00	6812.0	6.4	56.0
^{215}Po	1.00	7386.4	0.0	0.0
^{211}Pb	1.00	0.0	452.3	68.4
^{211}Bi	1.00	6550.0	9.9	46.7
^{211}Po	0.0027	20.3	0.0	0.0
^{207}Tl	0.9973	0.0	491.7	2.9
^{40}K	1.00	0.0	455.0	156.7
^{87}Rb	1.00	0.0	82.0	0.0

2. GAMMA SPECTROMETRY AND SECULAR EQUILIBRIUM

To find the radiation components D', at first glance the alpha spectrometry could be preferred because the major contributors to the dose of natural radiation are alpha particles (Bell, 1976, 1977). However in such a case the sample must be thin compared to the low alpha ranges to distinguish energies with satisfactory resolution. Contrary to the alpha particles, the gamma radiation penetrates matter very well. Therefore, the isotope concentration measurements by the gamma spectrometry are performed for large and thus more representative samples (Meakins *et al.*, 1978; Murray and Aitken, 1982; Zastawny and Rabsztyn, 1986; Debertin and Helmer, 1988).

In this way, the secular disequilibrium in the decay series is also checked and the source of disequilibrium can be located. The most frequent cause of disequilibrium is ^{222}Rn which, being a noble gas with a half-life of 3.8 days, may diffuse out of the sample. The next susceptible isotopes from uranium series are ^{238}U, ^{234}U and ^{226}Ra which form water soluble complexes and gaseous ^{220}Rn from the thorium series (Murray and Aitken, 1982; Krbetschek *et al.*, 1994; Olley *et al.*, 1997).

The gamma spectrometer Canberra System 100 applied in our laboratory consists of HPGe detector XtRa – GX1520. The detector and preamplifier are placed inside the low-background lead shield (model 747) and cooled by liquid nitrogen from vertical dipstick cryostat (7500SL). The integrated signal processor (model 1510) consists of a pulse height analysis system (Wilkinson ADC) to transform pulses which are finally collected by the computer-based MCA as a data file of gamma spectrum. Standard Marinelli beakers are used as sample containers for spectral measurements. Operating parameters of the system are governed and controlled by the computer programme – Canberra System MCA 100.

The following calibration tables have been established from measurements performed for Marinelli geometry and the standard mixture of radionuclides: the relationship between gamma energy and channel, the absolute efficiency calibration and the dependence of peak shape parameters on gamma energy. The spectrum of laboratory background activity has been established from prolonged measurements. The calibration and complete analysis of spectra in the range from 40 keV to 2 MeV are performed by analysis software-SAMPO90.

In the routine analysis of sample spectrum the fundamental algorithms of SAMPO (Koskelo *et al.*, 1981), i.e. peak search and fitting, multiplet deconvolution,

and background subtraction are supported by the calibration data. The table of calculated peak areas allows to perform nuclide identification and activity analysis. During this procedure the peak table is compared with the information located in the nuclide library file. The resulting nuclide activities are the least-squares best fit to the measured spectral data.

One of the most helpful features of SAMPO90 is its report generation capability. For TL dating, especially important section of the report is the nuclide identification list and the specific activity of isotopes (Bq/kg). When the gamma spectrum of a typical soil sample is measured during 100-200 h, the analysis allows one to find 11-17 activities of isotopes. Assuming the $^{87}Rb/^{40}K$ activity ratio of 0.142 (Warren, 1978), the effect of beta radiation from rubidium can also be included. **Table 2** shows an example of the activity report performed for a soil sample collected from the dune – Kępa Kujawska.

Table 2. *Radionuclide analysis, sample KĘPA 10B.*

Nuclide	Activity [Bq/kg]	Uncert. [%]	Error [Bq/kg]
^{228}Ac-Th	5.2	1.16	.060
^{212}Pb-Th	5.55	2.28	.127
^{212}Bi-Th	6.05	3	.181
^{208}Tl-Th	1.81	2.37	.043
^{234}Th-U	3.38	4.7	.159
^{226}Ra-U	3.69	18.16	.670
^{214}Pb-U	3.87	2.32	.090
^{214}Bi-U	5.69	1.54	.088
^{210}Pb-U	5.26	6.46	.340
^{235}U-Ac	.324	12.54	.041
^{40}K	476	4.84	23.038

In the case of disequilibrium in an old aeolian material it is likely that secular equilibrium will be found among two subsets consisting of pre-radon nuclides and radon daughters. Therefore, in this instance the isotopes from radioactive series can be divided into groups convenient for the dose rate calculation. Next the activity of the following groups of isotopes are considered:

1. Thorium series (full): ^{228}Ac, ^{212}Pb, ^{212}Bi, ^{208}Tl,
2. Pre-thoron: ^{228}Ac,
3. Pre-Rn ^{238}U series: ^{234}Th, ^{234}Pa(m), ^{226}Ra,
4. ^{222}Rn and after series: ^{214}Pb, ^{214}Bi, ^{210}Pb,
5. Uranium-235 series (full): ^{235}U, ^{223}Ra,
6. ^{40}K and ^{87}Rb.

From now on, the index j = 1,2...,6 will denote the respective group of isotopes.

Because of branching, the measured activity is multiplied by respective coefficient to obtain the parent activity (e.g. the activity of ^{208}Tl is multiplied by 2.786). Assuming the subset secular equilibrium, the weighted average activities, A(j) for the group j = 1, 3, 4 and 5 are calculated from the activities of members

$$A(j) = [\Sigma\, a(i) / \sigma^2(i)] / [\Sigma\, 1/\sigma^2(i)], \quad (3)$$

$$\sigma(j) = [\Sigma 1 / \sigma^2(i)]^{-1/2} . \quad (4)$$

In the case under discussion (KĘPA 10B) the averaged values are shown in **Table 3**. From now on, it is assumed that these averages describe each of the subset isotope from **Table 1**, even though it is missing in the activity report (**Table 2**).

Table 3. *Series activity, sample KĘPA 10B.*

Series	Activity [Bq/kg]	Error [Bq/kg]
^{232}Th full	5.284	0.0478
pre-thoron	5.2	0.060
^{238}U pre-Rn	3.396	0.155
Rn and after	4.817	0.006
^{235}U full	0.324	0.041
^{40}K	476.0	23.038
^{87}Rb	67.592	3.271

Referring to **Table 1** which gives the components of alpha, beta and gamma energy for each nuclide disintegration (i.e. including branching factor), the subset energies are calculated by summing up the energies of isotopes belonging to the defined chain and radiation. In Table 1 all the energies are given in keV and represent the energy release per parent disintegration. For TL dating purposes more convenient units are related to Bq, Gy and year.

Assuming 1 keV = $1.6022 \cdot 10^{-16}$ J, 1 a = 365.25 d = $3.1558 \cdot 10^{7}$ s, the annual doses data, R [μGy/a], defined for 1 Bq/kg, are collected in **Table 4**.

Table 4. *Annual dose for 1Bq/kg.*

Series	R_α [μGy/a]	R_β [μGy/a]	R_γ [μGy/a]
^{232}Th full	181.8	7.1	12.5
Pre-thoron	76.2	2.6	5.1
^{238}U full	216.8	11.3	9.1
Pre-^{222}Rn	93.1	4.3	0.2
^{222}Rn and after	123.8	7.0	8.9
^{235}U full	210.6	6.9	3.3
^{40}K+ ^{87}Rb	0.0	2.7	0.8

3. EVALUATION OF ANNUAL DOSE AND FINAL REPORT

The partial annual dose given in μGy/a for each radiation contribution is calculated as a product of the specific parent activity and a corresponding factor, R, from **Table 4**

$$D(j) = R(j)\, A(j). \quad (5)$$

According to data collected in **Table 3** and **Table 4**, the partial series doses for sample KĘPA10B are given in **Table 5**.

Table 5. *Series doses [μGy/a] for KĘPA 10B.*

Series	α [μGy/a]	β [μGy/a]	γ [μGy/a]
^{232}Th full	959.42	37.25	66.892
pre-thoron	395.72	13.10	23.296
^{238}U pre-Rn	314.69	15.42	.815
Rn and after	597.43	34.06	43.163
^{235}U full	68.39	1.97	.940
^{40}K	0	1094.8	376.04
^{87}Rb	0	27.0	0

In the case of subset of isotopes the averaged parent activity given by (3) is used. Of course, according to the kind of disequilibrium discovered, the appropriate groups and partial doses should be considered.

In general, the internal partial dose-rates are evaluated from measurements on dry material. As mentioned before, water also absorbs radiation. According to estimations performed for natural radioactivity by Zimmerman, the following equations and parameters are recommended by A. J. Aitken (1985) to include the effect of moisture into dating procedure:

$$D'(\alpha) = D''(\alpha) / (1 + 1.50\ W\ F), \quad (6)$$

$$D'(\beta) = D''(\beta) / (1 + 1.25\ W\ F), \quad (7)$$

$$D'(\gamma) = D''(\gamma) / (1 + 1.14\ W1\ F), \quad (8)$$

where D'' are the partial doses calculated from measurements on dry material, W and W1 are the saturation contents (weight of water/dry weight) for sample and soil respectively, F is the fraction uptake (the assumed average water content expressed as percentage).

The procedure of matrix dose calculation is completed with the estimation of the cosmic gamma dose, c(γ). For the samples which were buried at depth h≥2 m the following equation is applied (Aitken, 1985; Prescott and Hutton, 1994):

$$c = 178 \exp(-0.1715\ h), \quad (9)$$

where c is given in μGy/a. At shallower depth the dose is assumed to be 280 μGy/a for h ≤ 1 m and 150 μGy/a for 1 m < h < 2 m.

The partial doses are very flexible in applications. Depending on particular conditions of TL dating, defined by mineral composition, water contents, grain size fraction, extraction of the particular minerals and etching, specific coefficients k(α), k(β) and k(γ) may be used to determine «effective» dose- D', which is responsible for the natural TL. Algorithms described here are incorporated into the computer programme-EqDose, which is used for the TL dating purposes in our laboratory.

Results of calculations performed for sample KĘPA10b are summarised in the following final report:

KEPA10B	TOTAL Doses	sigma ERRORS
μGy/a	*μGy/a*	
alpha	1939.9	20.3
beta	1210.5	53.0
gamma	487.8	18.2
Cosm (2.7m)	112.1	0

TOTAL external radiation dose and sigma (μGy/a)
3750.396 +/- 59.622
Quartz-inclusion (μGy/a, alpha=0;
Etching and moisture effect)
1625.165 +/- 50.597
Fine-grain (μGy/a, TL efficiency and moisture effect)
1884.976 +/- 50.670
For:
alpha efficiency = 0.15
beta etching factor = 1
fraction of saturation; F = 0.8
sample saturation content; W = 0.1
soil saturation content; W1 = 0.2

The advantage of the presented method is that the partial matrix doses for alpha, beta and gamma radiation are determined directly and simultaneously from gamma spectrometry measurements.

Table 6. *Matrix doses (Kępa and Łążyn sites).*

No	Sample	Depth [m]	Total dose [μGy/a]
1	KEPA6B	1.1	4114
2	KEPA3B	1.1	4631
3	KEPA4B	1.4	3989
4	KEPA9B	1.4	3063
5	KEPA2B	2.7	3192
6	KEPA10B	2.7	3750
7	KEPA7B	3.9	3602
8	KEPA1B	5.5	3907
9	KEPA5B	7.0	4018
10	KEPA8B	7.0	3984
11	KEPA11B	7.0	3274
1	LAZ1	5.1	9961
2	LAZ2	5.2	14610
3	LAZ3	5.3	9246
4	LAZ4B	5.4	15251
5	LAZ5	5.5	6446
6	LAZ50B	5.5	9291
7	L97P1	7.0	2787
8	LAZB	7.5	2866
9	LAZC	9.5	2560
10	L97P2	11.0	2607
11	L51	12.6	9035
12	LAZD	13.5	4760
13	LAZYN4	17.0	2563
14	LAZYN3	17.5	3137
15	LAZYN2	18.0	3275
16	LAZYN1	18.1	3115

Finally, it should be stressed that in some place the strength of radioactivity sources differs substantially between various sedimentation layers of matrix. In such an environment, the radiation field is spatially inhomogenous and the resulting variability in dose rate cannot be exactly calculated, Hence, in selecting the location from which sample is to be extracted, emphasis should be given to avoid layers showing serious non-uniformity in radioactivity. When possible, preliminary measurements on pilot samples collected from the representative deposited layers should be made.

Table 6 shows the preliminary results of matrix dose measurements performed for samples from eolian (Kępa) and glacial-related (Łążyn) deposits. It is immediately apparent that the material deposited in Łążyn is heterogeneous, even within 0.1 m distance between different layers.

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