GAMMA SPECTROMETRY FOR OSL AND TL DATING OF LOESS DEPOSITS AT DYBAWKA AND TARNAWCE (SE POLAND)

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Key words: GAMMA SPECTROMETRY, NATURAL RADIOACTIVITY, INTERLABORATORY COMPARISON

Abstract: This article presents a comparison of results from the activity measurements of loess samples obtained with two methods in two laboratories. The methods are semiconductor gamma radiation spectrometry and gamma radiation scintillation spectrometry. Various measurement geometries and various sets of radioactivity standards were applied. Samples were collected from two loess profiles, namely Dybawka and Tarnawce. The results of activity measurements for natural radioisotopes of uranium, thorium and potassium obtained in the Gliwice and Gdańsk laboratories are in good agreement. Additionally, the activity of 235U was measured in the analysed samples.

1. INTRODUCTION

In addition to the radiocarbon dating method, optically stimulated luminescence (OSL) and thermoluminescence (TL) methods are at present widely used to determine the age of geological formations and archaeological objects (Bluszcz, 2001; Bluszcz et al., 2001; Prescott et al., 1997). In order to determine the age of a given sample with luminescence methods, it is necessary to know the equivalent dose (absorbed dose) and the annual dose (dose rate). The dose rate may be determined in a number of ways. The most frequent methods are TL dosimetry and gamma radiation spectrometry. TL dosimetry involves the use special dosemeters made of LiF, CaF₂, or Al₂O₃ (Bluszcz, 2000). The dosemeters are placed directly in the analysed deposit for a precisely specified time, but not shorter than 3 months (Bluszcz, 2000). The dose rate measurement with the use of the above mentioned dosemeters is a long-term process with a relatively high uncertainty (a dozen or so). Several years ago, LiF dosimeters were used in the Gdańsk laboratory for dose rate in situ measurements (Fedorowicz, 1990 and 2003). A dose rate may be also determined with the alpha radiation spectrometry method. However, despite the fact that this method allows the determination of activity in radioactive series, it is not possible to specify the activity of 40K potassium in the sample (Bluszcz, 2000).

To determine the dose rate the gamma radiation spectrometry method may be used. This method allows a relatively easy determination of the natural activity in sample. The activities of 238U, 235U and 232Th series can be determined and also the activity of 40K. Compared to alpha radiation spectrometry, the important advantage of this method is the lack of tedious chemical extraction. Gamma radiation semiconductor spectrometry has the width of a photopeak of about 2 keV, compared to 50 keV for gamma radiation scintillation spectrometry. Thanks to a better energy resolution, it is possible to measure an isotope activity in more detail despite the presence of the Compton continuum, and to specify the degree of disequilibrium in the radioactive series due to the possibility of determination of the activity of respective elements of radioactive series.

In respect to luminescence dating methods, in practice it is assumed that isotopes of uranium and thorium series are in radioactive equilibrium. Radioactive equilibrium disturbances are rather small in thorium series; however, significant disturbances of equilibrium may be noted in the case of uranium series (Olley et al., 1996). The main source of disturbance is the escape of 222Rn, which may escape outside. Another source of disequilibrium in the uranium series is a relatively high mobility of easily dissoluble compounds of 234U and 238U (Hercman, 1999; Oczkowski et al., 2001).

Regardless of the fact which measurement method is selected, the specification of radioisotope activities is subject to measurement uncertainty. If such activities are specified with a gamma radiation spectrometry method,
uncertainty is mainly related to the uncertainty of radioactivity determination and also to the uncertainty of radioactive standards and to the uncertainty of the background counting.

In order to obtain more precise results of isotope activity measurements in the samples to be dated with luminescence techniques, two laboratories: the Gdańsk laboratory from the Department of Geomorphology and Quaternary Geology at the Gdańsk University and the Luminescence Dating Laboratory in the Department of Radioisotopes in the Institute of Physics at the Silesian University of Technology in Gliwice, conducted interlaboratory comparative measurements of samples dated with the OSL and TL methods. For purposes of this study, the laboratories measured the natural radioactivity of samples from two different geological profiles: Dybawka and Tarnawce. The measurements were carried out with two various gamma radioactivity determination techniques and with the application of various radioactivity standards and various measurement geometries.

2. MATERIAL AND METHODS

The following methods were applied: gamma radiation scintillation spectrometry and gamma radiation semiconductor spectrometry. In the case of gamma radiation scintillation spectrometry, the following devices were used: MAZAR-95 and TUKAN. Both MAZAR and TUKAN have NaJ/Tl scintillation detector type SSU 70. MAZAR is a three-channel scintillation spectrometer, which automatically converts counts in the $^{40}\text{K}$, $^{226}\text{Ra}$, $^{228}\text{Th}$ channels into concentrations of such elements and alpha, beta, gamma radiation doses. The TUKAN scintillation spectrometer has 4096 channels. Its analog-to-digital converter has a 12-bit resolution. It conducts compensatory conversions with the 10-bit averaging of channel width. Marinelli containers with a capacity of 0.5 dm$^3$ (a cylinder with $r=0.4$ dm, $h=1$ dm) were used as measurement containers. For purposes of gamma radiation semiconductor spectrometry, a coaxial germanium detector of high purity (HPGe) CANBERA type P and of 55% efficiency was used. In this case, samples were measured with the application of two various measurement geometries: Marinelli with a capacity of 0.6 dm$^3$ and the medium box of 130 cm$^3$.

Three sets of radioactive standards prepared by CLOR (Central Laboratory for Radiological Protection in Warsaw, Poland) were used as calibration standards. The two sets of standards were used for the measurements with the semiconductor spectrometer, separately for each measurement geometry, and one set of standards was used for the scintillator spectrometers. The measurement time was different for various detectors and ranged from 25,000 to 86,000 s. In the case of MAZAR-95 system, twenty measurements of 2,000 s each were conducted. Prior to the measurements, all the samples were dried in the laboratory dryer and sifted, and then placed in the containers, tightly closed, and seasoned for the period of 30 days to obtain an equilibrium in radioactive series.

Comparative activity measurements were conducted for 9 samples from the two geological profiles: Dybawka (DA) and Tarnawce (TA). Both profiles are located in the vicinity of Przemyśl (SE Poland). The samples analysed are loess formations (Lanczont and Fedorowicz, 2004). Both loess profiles are in close proximity at a distance of approx. 2 km. The TA and DA profiles are outcrops of approx. 20 m thickness. The stratigraphy of the profiles was determined on the basis of their geological examination (Lanczont, 1991). Profiles differ with the time of sediment deposition. Dybawka loess is Vistulian (Upper Pleniglacial), while Tarnawce loess shows a larger time span (from Vistulian loess to Wartanian loess).

Table 1 presents sample weights for various measurement containers and techniques applied. Table 2 shows the activity of radioactive standards used in the experiment. The isotope concentration of natural radioactive series $^{238}\text{U}$, $^{232}\text{Th}$ and the concentration of $^{40}\text{K}$ were measured. The activity of $^{235}\text{U}$ isotope was also specified in the samples covered by the analysis.

| Table 1. Masses of samples used in the experiment. |
| Sample name | Mass for the Marinelli container, HPGe detector (g) | Mass for the Medium container, HPGe detector (g) | Mass for the Marinelli container, scintillator detectors (g) |
| DA-2 | 806.9 | 162.72 | 621.4 |
| DA-5 | 717.2 | 168.02 | 548.6 |
| DA-6 | 752.7 | 168.90 | 609.4 |
| DA-9 | 853.7 | 179.41 | 612.4 |
| TA-1 | 838.0 | 163.04 | 613.4 |
| TA-2 | 679.7 | 171.21 | 496.4 |
| TA-4 | 816.1 | 160.72 | 642.4 |
| TA-14 | 832.4 | 169.96 | 603.3 |
| TA-16 | 825.6 | 175.72 | 625.5 |

| Table 2. Activity of radioactive standards used in the experiment (in Bq/kg). |
| Isotope | Detector |
| $^{226}\text{Ra}$ | $^{228}\text{Th}$ | $^{40}\text{K}$ |
| HPGe, Marinelli container | HPGe, Medium container | Scintillator detector |
| Detector | 2,970 | 2,760 |
| Detector | 1,016 | 939 |
| Detector | 15,300 | 15,823 |
3. CALCULATION OF ACTIVITY

In the case of gamma radiation scintillation spectrometry, the activity of deposit samples was measured in three intervals of gamma radiation photon energy. Photons from the $^{40}\text{K}$ potassium isotope of 1.46 MeV energy were registered in the first interval (1.26-1.65 MeV); photons from the $^{214}\text{Bi}$ isotope of 1.76 MeV were registered in the second interval (1.65-2.30 MeV) and photons from the $^{208}\text{TI}$ isotope of 2.62 MeV energy were registered in the third interval. The $^{214}\text{Bi}$ and $^{208}\text{TI}$ activities that were specified may be adopted as activities in the uranium and thorium series if they are in secular equilibrium.

In contrast to the scintillator method, the gamma radiation semiconductor spectrometry allows one to specify the activity of a larger number of radioactive isotopes. In respect of uranium series, it is not possible to determine the $^{238}\text{U}$ activity because this isotope has a line of a very small efficiency 0.064% of a relatively low energy of 49.6 keV (Papachristodoulou et al., 2003). In addition, it is located relatively close to the line of the $^{210}\text{Pb}$ isotope with the energy of $E_G=46.5$ keV and efficiency of 4%. In order to specify the activity of $^{238}\text{U}$ series, the 1001 keV line from the $^{234}\text{Pa}$ isotope may be used (Yucel et al., 1998). The concentration of $^{226}\text{Ra}$ was determined with the line of energy equal to 186.1 keV. The $^{226}\text{Ra}$ daughters activity were determined with the use of the following lines: 295.2 keV (19.2% efficiency), and 352 keV (37.1% efficiency) from $^{214}\text{Pb}$ and lines 609 keV (46% efficiency), 1120 keV (15% efficiency), and 1764.5 keV (15.9% efficiency) from $^{214}\text{Bi}$.

The activity in the $^{232}\text{Th}$ thorium series was specified with the use of $^{238}\text{Ac}$ lines (with energy of 338 and 911 keV and efficiencies of 11.3% and 26.6%, respectively), the radioactive equilibrium between $^{238}\text{Ac}$ and $^{232}\text{Th}$ being assumed. The activity in the $^{232}\text{Th}$ series after $^{220}\text{Rn}$ was determined with the use of $^{208}\text{TI}$ lines (with energy of 583 and 2614 keV and efficiency of 30.6 and 35.8%, respectively).

Furthermore, the activity of $^{235}\text{U}$ was determined. This isotope has several lines of gamma radiation: 143.8 keV, 163.3 keV, 187.5 keV, and 205.3 keV with the efficiency of 10.5%, 4.7%, 53.0%, and 4.7%, respectively. Besides, the $^{227}\text{Th}$ isotope of this series has lines with energy 93.9 keV and 236.0 keV, but of relatively low efficiency. In respect of $^{219}\text{Rn}$ daughter elements, a relatively efficient line of gamma radiation is the line derived from $^{211}\text{Bi}$, with the energy of 351.1 keV and the efficiency of 12.9%. It lies close to the line from $^{210}\text{Pb}$, with the energy of 352.0 keV and efficiency of 37.1%.

The activity of $^{40}\text{K}$ isotope in the samples was determined with the use of the 1.460 keV line with the efficiency of 10.7%. It is relatively distant from the other lines, and in particular from the intensive lines of uranium and thorium series isotopes.

4. RESULTS AND DISCUSSION

Figs 1-3 present results of activity measurements in the $^{238}\text{U}$ and $^{232}\text{Th}$ series, and of $^{40}\text{K}$ in soil samples as conducted by the two laboratories. Table 3 presents the activities of the $^{235}\text{U}$ series isotopes.

### Table 3. The results of $^{235}\text{U}$ activity with the use of HPGe detector (Bq/kg).

<table>
<thead>
<tr>
<th>Sample name</th>
<th>Depth (m)</th>
<th>Activity, incl. uncertainty (Bq/kg)</th>
<th>$^{235}\text{U}/^{238}\text{U}$ Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>DA-2</td>
<td>2.25</td>
<td>1.31±0.24</td>
<td>31.1</td>
</tr>
<tr>
<td>DA-3</td>
<td>3.45</td>
<td>1.28±0.33</td>
<td>31.6</td>
</tr>
<tr>
<td>DA-4</td>
<td>5.15</td>
<td>1.53±0.31</td>
<td>25.3</td>
</tr>
<tr>
<td>DA-9</td>
<td>7.95</td>
<td>1.44±0.25</td>
<td>24.9</td>
</tr>
<tr>
<td>TA-1</td>
<td>1.60</td>
<td>1.13±0.30</td>
<td>31.4</td>
</tr>
<tr>
<td>TA-2</td>
<td>2.80</td>
<td>1.25±0.36</td>
<td>30.0</td>
</tr>
<tr>
<td>TA-4</td>
<td>3.70</td>
<td>1.34±0.33</td>
<td>23.5</td>
</tr>
<tr>
<td>TA-14</td>
<td>10.50</td>
<td>1.14±0.29</td>
<td>25.1</td>
</tr>
<tr>
<td>TA-16</td>
<td>12.60</td>
<td>1.11±0.32</td>
<td>31.8</td>
</tr>
</tbody>
</table>

### Table 4. Comparison of results of activity measurements in $^{238}\text{U}$ series.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Depth (m)</th>
<th>All laboratories</th>
<th>Weighted mean (Bq/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Semiconductor detectors</td>
<td></td>
</tr>
<tr>
<td>DA-2</td>
<td>2.25</td>
<td>39.71±0.82</td>
<td>39.67±0.87</td>
</tr>
<tr>
<td>DA-5</td>
<td>3.45</td>
<td>38.11±0.85</td>
<td>38.60±0.91</td>
</tr>
<tr>
<td>DA-6</td>
<td>5.15</td>
<td>37.13±0.79</td>
<td>37.38±0.85</td>
</tr>
<tr>
<td>DA-9</td>
<td>7.95</td>
<td>35.02±0.68</td>
<td>35.41±0.77</td>
</tr>
<tr>
<td>TA-1</td>
<td>1.60</td>
<td>34.61±0.72</td>
<td>34.41±0.77</td>
</tr>
<tr>
<td>TA-2</td>
<td>2.80</td>
<td>36.98±0.87</td>
<td>37.33±0.92</td>
</tr>
<tr>
<td>TA-4</td>
<td>3.70</td>
<td>31.84±0.70</td>
<td>31.64±0.74</td>
</tr>
<tr>
<td>TA-14</td>
<td>10.50</td>
<td>28.65±0.67</td>
<td>28.57±0.71</td>
</tr>
<tr>
<td>TA-16</td>
<td>12.60</td>
<td>33.24±0.76</td>
<td>33.62±0.83</td>
</tr>
</tbody>
</table>
Table 5. Comparison of results of activity measurements in 232Th series.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Depth (m)</th>
<th>All laboratories</th>
<th>Weighted mean (Bq/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>Semiconductor detectors</td>
</tr>
<tr>
<td>DA-2</td>
<td>2.25</td>
<td>40.62±0.78</td>
<td>40.72±0.82</td>
</tr>
<tr>
<td>DA-5</td>
<td>3.45</td>
<td>37.30±0.83</td>
<td>37.21±0.87</td>
</tr>
<tr>
<td>DA-6</td>
<td>5.15</td>
<td>36.14±0.77</td>
<td>36.25±0.83</td>
</tr>
<tr>
<td>DA-9</td>
<td>7.95</td>
<td>33.78±0.67</td>
<td>33.84±0.70</td>
</tr>
<tr>
<td>TA-1</td>
<td>1.60</td>
<td>34.98±0.72</td>
<td>35.11±0.77</td>
</tr>
<tr>
<td>TA-2</td>
<td>2.80</td>
<td>39.71±0.87</td>
<td>39.98±0.92</td>
</tr>
<tr>
<td>TA-4</td>
<td>3.70</td>
<td>41.23±0.85</td>
<td>41.40±0.89</td>
</tr>
<tr>
<td>TA-14</td>
<td>10.50</td>
<td>33.55±0.73</td>
<td>33.43±0.77</td>
</tr>
<tr>
<td>TA-16</td>
<td>12.60</td>
<td>36.28±0.79</td>
<td>36.40±0.84</td>
</tr>
</tbody>
</table>

Table 6. Comparison of results of 40K activity measurements.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Depth (m)</th>
<th>All laboratories</th>
<th>Weighted mean (Bq/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>Semiconductor detectors</td>
</tr>
<tr>
<td>DA-2</td>
<td>2.25</td>
<td>578.3±9.1</td>
<td>577±10</td>
</tr>
<tr>
<td>DA-5</td>
<td>3.45</td>
<td>516.7±8.6</td>
<td>518.7±9.6</td>
</tr>
<tr>
<td>DA-6</td>
<td>5.15</td>
<td>510.9±8.6</td>
<td>512.1±9.6</td>
</tr>
<tr>
<td>DA-9</td>
<td>7.95</td>
<td>484.5±7.5</td>
<td>483.8±8.3</td>
</tr>
<tr>
<td>TA-1</td>
<td>1.60</td>
<td>500.2±8.1</td>
<td>499.0±9.1</td>
</tr>
<tr>
<td>TA-2</td>
<td>2.80</td>
<td>561.2±8.8</td>
<td>565±10</td>
</tr>
<tr>
<td>TA-4</td>
<td>3.70</td>
<td>525.1±8.3</td>
<td>526.5±9.6</td>
</tr>
<tr>
<td>TA-14</td>
<td>10.50</td>
<td>525.4±8.5</td>
<td>526.8±9.6</td>
</tr>
<tr>
<td>TA-16</td>
<td>12.60</td>
<td>485.2±7.8</td>
<td>481.5±8.9</td>
</tr>
</tbody>
</table>
Comparison of results obtained with various measurement methods

Tables 4, 5, and 6 present the comparison of results obtained in the Gliwice laboratory with the results from the Gdańsk laboratory. These tables present weighted averages for all the measurements of the same sample and weighted averages calculated separately for both laboratories (various measurement techniques).

To compare the figures obtained in the two laboratories with different techniques, scaled deviations were computed for the activity obtained for respective radioisotopes in accordance with the following pattern:

$$Semi\text{-}Scin\text{ }dev = \frac{WM\text{ }sem - WM\text{ }scint}{\sqrt{\sigma^{2}_sem + \sigma^{2}_scint}}$$

where:

- $WM\text{ }sem$ – weighted mean for measurements obtained with the semiconductor detector,
- $WM\text{ }scint$ – weighted mean for measurements obtained with scintillator detectors,
- $\sigma^{2}_sem$ – standard deviation of weighted mean from measurements obtained with the semiconductor detector,
- $\sigma^{2}_scint$ – standard deviation of weighted mean from measurements obtained with the scintillator detector.

Table 7 presents scaled deviations between the figures obtained with the two measurement techniques.

The results obtained for two laboratories were compared by using the t-Student test. There are no significant differences between the results obtained in those laboratories.

The comparison of weighted averages of results obtained in the two different laboratories with the two various measurement methods shows no clear systematic differences between the results obtained in the two laboratories. Therefore, it may be concluded that there are no significant statistical differences between the results obtained by the two laboratories.

Discussion of the Activity of Dybawka and Tarnawce Samples

The $^{232}$Th isotope series in the samples analysed ranges from $33.55 \pm 0.73$ Bq/kg to $41.25 \pm 0.85$ Bq/kg (the values of weighted averages of all the values). It is larger than the average value for the area of Poland, which amounts to $20.7$ Bq/kg (Jagielak et al., 1998). It may be observed that in respect of the DA series, thorium series isotopes decrease in proportion to depth while no changes of activity in relation to depth were observed in the TA series.

The $^{40}$K content ranges from $484.5 \pm 7.5$ Bq/kg to $578.3 \pm 9.1$ Bq/kg in the samples analysed and it is slightly higher than Poland’s average which amounts to $410$ Bq/kg (Jagielak et al., 1998).

The $^{40}$K content in the DA series samples shows a declining tendency in relation to the increase of depth. In respect of TA series, this tendency may also be observed, starting from the TA-2 sample.

Fedorowicz S., 1999: Z datowań TL wierzutlianu nad Wistłą. (From the dating of Vistulian TL along the Vistula) Peribalticum 6: 17–44.


