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GAMMA BACKGROUND MEASUREMENTS BY TL METHOD: APPLICATIONS IN LOCATIONS WITH VARIED GEOLOGICAL CONTEXT

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Abstract: The aim of this study was to establish whether the thermoluminescence (TL) method is suitable for natural gamma dose monitoring with the ultimate goal to apply this method in the project “Radon map for Center, West and North West regions from Romania” that has the additional aim of establishing a high resolution database for natural gamma dose rates. Seven locations with varied geological context have been selected (Arad, Cluj-Napoca, Bologa, Miercurea-Ciuc, Predeal, Belis, Sinaia). Ultrasensitive TL dosimeters, LiF:Mg,Cu,P were used in order to quantify the gamma doses. The calibration was performed using a gamma ^{60}Co source. The dose response was linear, with a determination coefficient of $R^2 = 0.96$ for MCP-N detectors and 0.99 for MCP-7 type. For MCP-N dosimeters, the sensitivity was 0.375±0.040 µC/mGy, while for MCP-7 dosimeters the determined sensitivity was 0.363±0.018 µC/mGy for our dosimetric system (Harshaw 3500 TLD Reader). For Belis location, gamma dose rate was found to be 133±16 nGy/h for MCP-N detectors and 158±8 nGy/h for MCP-7, higher than in the other locations (Arad:93±10 nGy/h and 115±7 nGy/h; Cluj:102±12 nGy/h respectively 110±7 nGy/h; Bologa:102±11 nGy/h and 114±8 nGy/h; Miercurea-Ciuc:105±12 nGy/h respectively 116±6 nGy/h; Predeal:95±10 nGy/h and 111±7 nGy/h; Sinaia:87±10 nGy/h and 99±5 nGy/h). The results can be explained by the existent geological substrate in this area, a granitic one, with rocks rich in ^{235}U and ^{40}K. The feasibility test of the investigated method will successfully serve for obtaining realistic input data for the effective dose received by the population.

Keywords: gamma background, thermoluminescence dosimetry, geological substrate, dose rates, dosimeter

1. INTRODUCTION

Humans, as all organisms, have always been exposed to natural ionising radiations (Vandercasteele, 2004). Natural radioactivity is strongly influenced by cosmic radiation and terrestrial sources. The contribution of cosmic rays to the natural radioactivity depends on geographic position, altitude and solar activity. The main contribution to external exposure is given by gamma emitting radionuclides from the soil such as ^{238}U, ^{232}Th series and ^{40}K, which are associated with minerals such as monazite and zircon (Jayanthi et al., 2011, Mishev & Hristova, 2012). Other significant contribution is due to radon gas and its radioactive progeny (UNSCEAR 2008). This gas is released from the Earth’s crust and subsequently decays into radioactive atoms. Radon is emanated from soil, rock, sediments and water and causes respiratory diseases, especially lung cancer when having a prolonged exposure (Cosma et al., 2013). The recent pooled analysis of key European studies showed that 9% of deaths from lung cancer per year in Europe were estimated to be attributable to exposure to indoor radon (Darby et al., 2006). Also, radioactive atoms produced in the upper atmosphere by high-energy cosmic rays contribute to the exposure. Man-made sources give 3% of the background radiation. The level of natural radioactivity varies from one location to another over the world with some areas where the level is significantly higher than the average (Hendry et al., 2009).

Environmental monitoring of radiation doses using passive thermoluminescent dosimeters has found more and more applications in the last 35 years (Olko et al., 2004). This type of monitor allows one to measure the long-term accumulation of dose at different stations distributed in the environment (Taam et al., 2008). In 2000, the Romanian National Commission for Nuclear Activities Control has limited public dose to 1 mSv/year above the natural background (dictate no. 14 /2000) according to the Basic Safety Standards and ICRP 60 recommendations (IAEA, 1996, ICRP, 1991). Therefore, more sensitive dosimeters with lower detection limit are required in order to comply with this new public dose limit and LiF:Mg,Cu,P is capable to measure such low doses. This material is one of the most attractive thermoluminescent one used for assessment of low radiation doses due to its great dosimetric characteristics such as ultra-high sensitivity, long-term information storage, easy handling, simple readout, good reproducibility, negligible fading, good linearity (1 µGy – 10 Gy) and tissue equivalence (Imatoukene et al., 2008; Olko, 2010).

In the recent years, many scientists investigated natural radioactivity with interest for gamma background measurements (Budzanowski et al., 2004, Cosma et al., 2008, Almgren et al., 2008a, Almgren et al., 2008b, Stochioiu et al., 2008, Trevisi et al, 2010, Evangelista et al., 2012, Mishev & Hristova, 2012, Anjos et al., 2010, Jayanthi et al., 2011, Taam et al., 2008, Saidou et al., 2010). In all cases the main conclusion was that doses for natural radioactivity have important variations from one location to another and it is crucial to have a complete database for natural gamma dose rates.

The aim of this paper was to establish that thermoluminescence (TL) method is suitable for natural gamma dose monitoring with the ultimate goal to apply this method for “Radon map (residential, geogenic, water) for Center, West and Northwest regions from Romania” research project. The importance of these measurements is two–fold: (i) outdoor gamma dose rates obtained will allow establishing a very high resolution database for natural gamma dose rates in the investigated region that can be integrated into the seasonal maps of natural terrestrial dose rates at the European level (e.g. Szegvary et al., 2007), (ii) the corroboration of indoor radon concentration levels and indoor as well as outdoor gamma dose rates can serve for realistic input data for the effective dose received by the population in the investigated area to be computed (UNSCEAR 2000).

2. SELECTED LOCATIONS

Arad is a city situated on the right bank of the Mures river in the Banat region of western Romania. The geology of the area is characterized by up to 100 m thick Holocene and Pleistocene sediments (sands, gravels, clays, and loess) underlying Neogene sedimentary units composed by sands and clays having up to 3000 m thick (Mutihac, 1990). Located in the Somesul Mic river valley, Cluj-Napoca metropolitan area lies at the confluence of the Apuseni mountains, the Someş plateau and the Transylvanian plain. Substrate geology of this region is represented by holocene alluvial deposits (Transylvanian Basin) formed by sands, gravels underlying Upper Eocene-Lower Oligocene marine and continental deposits (limestones, marls, sandstones, clays, and gypsum) (Baciu & Filipescu, 2002). Bologa village is situated at the confluence of the Crişul Repede and Henţ (Sebeş or Săcuieu) rivers (the confluence is known as "gura apelor" - "the mouth of the waters" - in the local toponymy) and at the foot of the Vlădeasa mountains (1863 m), part of the Apuseni Carpathians. In this region there is evidence of the presence of paleocene dacites and andesites (North Apuseni Mountains) locally covered by thin alluvial deposits (sands and gravels) along the streams (Ianovici et al., 1976). Miercurea Ciuc city is situated at an altitude of 662 m, on the river Olt, in Ciuc depression. The geology of the area is characterized by up to 40 m thick Holocene and Pleistocene sediments (clays, sands, gravels, and loess) deposited in an intramontane basin underlying Lower-Middle Pleistocene pyroxene and amphiboles andesites (eastern Carpathians) (Mutihac, 1990). Predeal is situated in the Prahova Valley, in the southern part of Brașov county. Neighboring towns include Azuga to the south, Bușteni to the southwest, Râșnov to the northwest and Brașov to the north. The town is located in a mountainous area, with the Piatra Mare mountains to the north, the Bucegi mountains to the southwest and the Postăvarul massif to the northwest. Another town considered in this study is Sinaia which is also a mountain resort in Prahova county, Romania. It is located at about 60 km northwest of Ploiești and 50 km south of Brașov, in a mountainous area on the Prahova River valley, just east of the Bucegi Mountains. The altitude of the town varies between 767 m and 860 m. From a geological viewpoint, the areas are characterized by the presence of the lower Cretaceous flysch deposits (Eastern Carpathians) made by sandstones, shales, and limestones (Mutihac, 1990). Belis village is extended north-west slopes of the mountains Gilău and south-southeast of Magura Călăţele Apuseni mountains, on the banks of the anthropogenic Fântânele Lake of the upper Warm Somes river. The village is situated at an altitude of 1,050m. The geology of the area is characterized by the existence of upper Paleozoic granitoid rocks of Muntele Mare magmatic body intruded in metamorphic rocks (North Apuseni Mountains) (Balintoni, 1997).

3. MATERIALS AND METHODS

In order to measure gamma dose rates, ultra-sensitive LiF:Mg,Cu,P dosimeters (code MCP-N and MCP-7 produced by TLD Poland) were used, in the form of 4.5 mm diameter and 0.9 mm thickness pellets. The standard annealing procedure by heating the dosimeters at 240°C for 10 minutes was performed before placing the dosimeters in the environment. Seven sets of dosimeters (at least 2 MCP-N pellets and at least 3 MCP-7 pellets for each location) were packed in closed plastic bags and coded. Every detector was wrapped in aluminum foil in order to have humidity and light protection and placed at a distance of 1 meter above ground, according to standard procedures (Olko, 2004). The time exposure was set to at least 3 weeks. After this period, TLDs were collected and brought into the laboratory and read with a Harshaw 3500 TLD Reader. The time temperature profile (TTP) for dosimeters reading was selected by having four regions of interest in the glow curve (ROI 1: first 50 channels, ROI 2: channel 50 to channel 120, ROI 3: channel 120 to channel 155 and ROI 4 channel 155 to channel 200). The heating rate was 5°C/s with the maximum temperature of 220°C (for 20 seconds). The third region of interest has been used for integration, as in this region the main peak at 210°C of LiF:Mg,Cu,P is registered. Calibration was performed using a ^{60}Co radioactive source from Physics Faculty, Babes-Bolyai University. Background subtraction was performed using the response of a batch of freshly annealed dosimeters.

4. RESULTS AND DISCUSSION

Background signal was determined for both types of detectors and was found to be equal to 0.29 nC for MPC-N type (see Figure 1a) and 0.32 nC for MCP-7 type (see Figure 1b). The lowest limit of detection for our system can be estimated as a ratio between there times the standard deviation of background signal (nC) and the sensitivity of the dosimeters (μC/mGy) and was found to be equal to 304 nGy for MCP-N detectors and 175 nGy for MCP-7 detectors. This is a very realistic value taking into account the lowest detectable dose for these detectors is around 60 nGy (Olko, 2004).

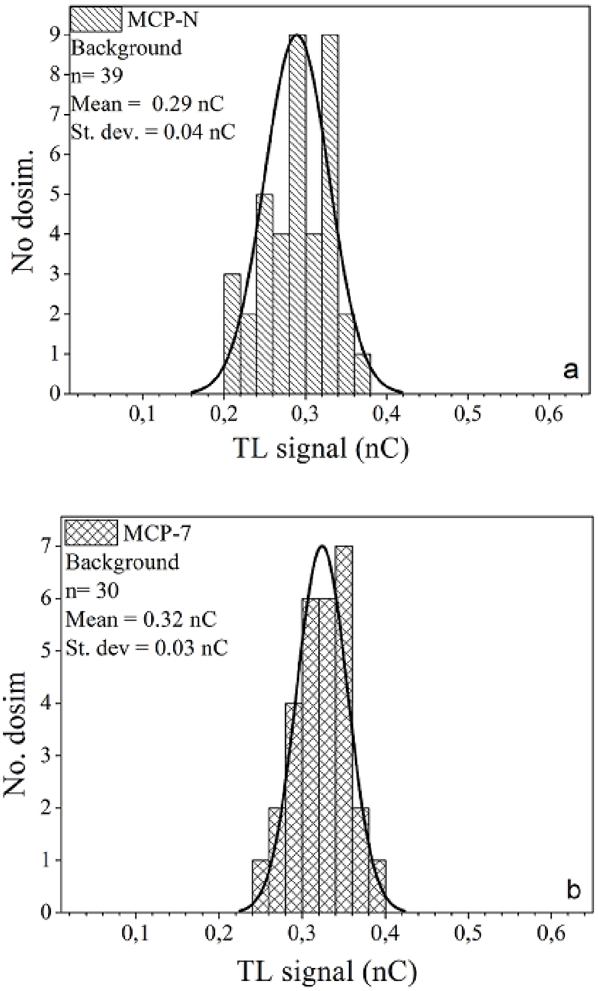

Figure 1. Histogram of background signal for MCP-N (a), respectively MCP-7(b) detectors. It can be observed that the distribution for the batch of detectors is normal and background signals are very small (nC).

Figure 3 presents a typical glow curve for a MCP-N detector exposed at Belis. The integral TL response in the region of interest is 57 nC. It can be noticed that this value is 2 orders of magnitude higher than the background recorded for this type of detector. This result confirms that the TL method using MCP detectors is ultra-sensitive and can be applied for monitoring environments with low dose rates (tens of nGy/h) for exposure times of less than a few weeks.

The determined gamma dose rates for the investigated locations are presented in table 1 and plotted for the sake of clarity in figure 4, where these values are compared to the gamma dose rate information available at the moment for Romania in the UNSCEAR 2008 report. Except Belis location, for all other locations, results were very close to the Romanian average which is 92 nGy/h.

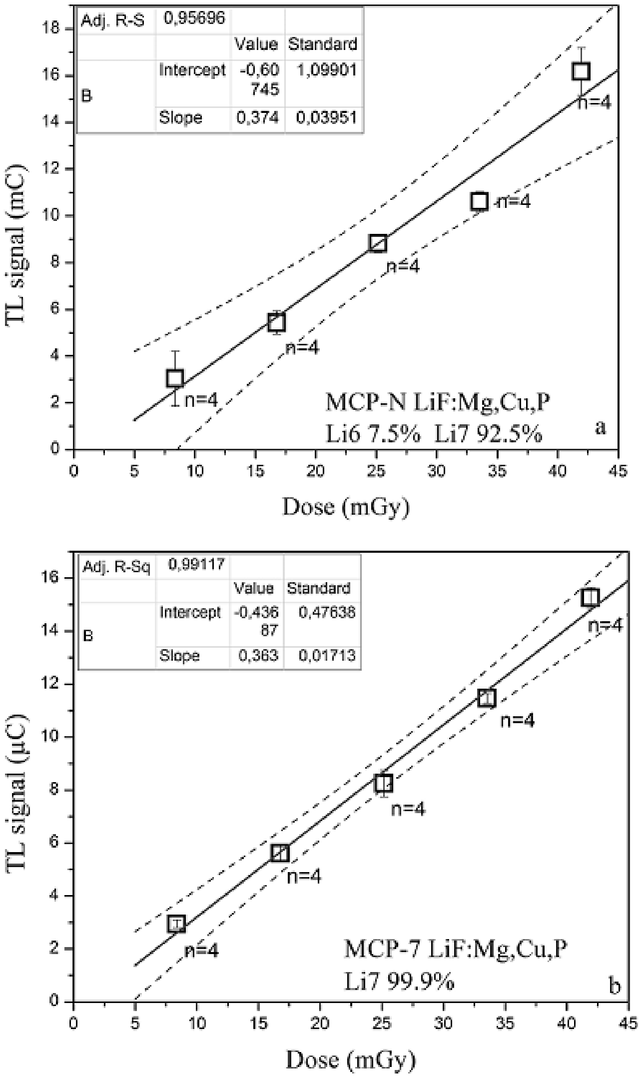

Figure 2. Calibration of MCP-N and MCP-7 TL detectors respectively. The number of detectors used for each dose point is denoted in the graph. It can be noted that the dose response to ^{60}Co is highly linear (solid line).Dashed lines represent the 95% confidence limits.

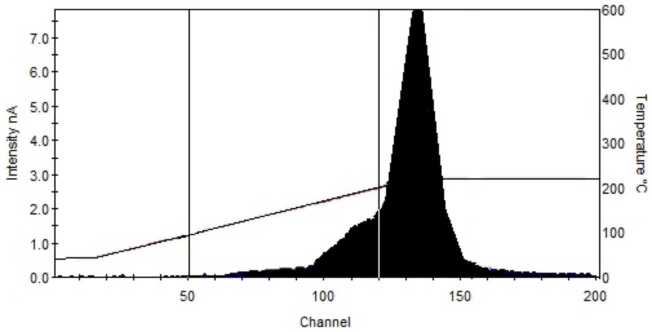

Figure 3. Typical glow curve for Belis location. The dosimetric peak at 210 ^{o}C can be observed in the third selected region of interest.

It can be noted that the dose rates were in the normal range of variation given by UNSCEAR Report 2008, which is 52-163 nGy/h. The higher dose rate observed for Belis location (133±16 nGy/h for MCP-N detectors and 158±8 nGy/h for MCP-7 type) is explained by the existent geological substrate in this area, a granitic one, with rocks rich in uranium and potassium that increase the existing natural background (Moldovan et al., 2013). The good agreement between the dose rates determined using MCP-N and MCP-7 detectors further confirms the robustness of the applied method.

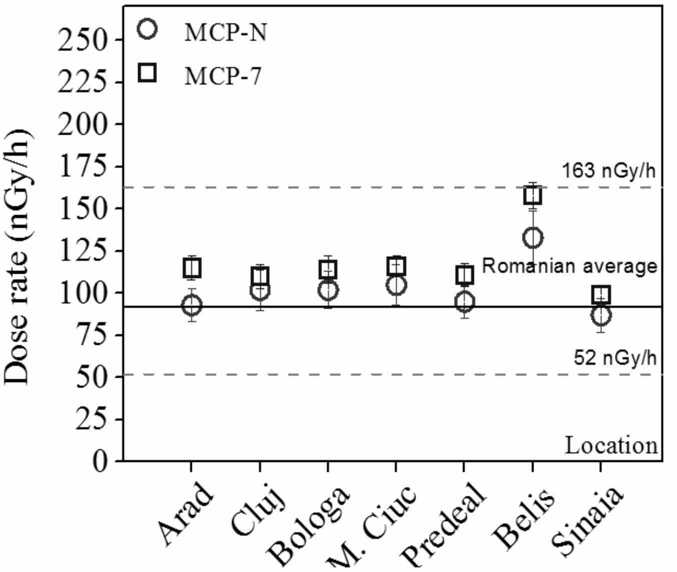

Figure 4. Gamma dose rates obtained for the 7 locations considered. The continous line represents the Romanian average for gamma dose rates – 92 nGy/h (UNSCEAR Report 2008). The two dashed lines represent the interval of variation for gamma dose rates in Romania – 52-163 nGy/h (UNSCEAR Report 2008).

5. CONCLUSIONS

Highly sensitive MCPs (LiF:Mg,Cu,P) thermoluminescent detectors were applied for environmental monitoring of radiation doses. Due to the high sensitivity of MCP detectors, our dosimetric system allows for short term (a couple of weeks) integrated measurements to be performed, as well as long term (months and up to years) surveys of environmental doses to be carried out. The lowest gamma dose rates have been recorded in a location characterized by sedimentary rocks (Sinaia, Predeal) and thick sediments units (Arad). Alluvial deposits (Cluj), thin sediments layers (Miercurea Ciuc) and volcanic rocks (Bologa) show lower to medium values. One single location (Beliş) indicates a very strong correlation between geological setting and gamma dose rates: the highest values are due to the presence of granitoid rocks composed by radioisotopes bearing minerals (feldspars and other silicates).

Table 1. TL signal, gamma dose and gamma dose rate obtained for each investigated location

Location	Time of exposure (days)	Type of dosimeters	Mean signal (nC)	Bck. corrected signal (nC)	Signal/day (nC)	Gamma dose/day (μGy)	Gamma dose rate (nGy/h)	Geological setting
Arad	24	mcp-n	20.34	20.05	0.83	2.23±0.24	93±10	Sediments
		mcp-7	24.42	24.10	1	2.77±0.17	115±7	
Cluj	40	mcp-n	36.96	36.67	0.92	2.44±0.30	102±12	Alluvial deposits
		mcp-7	38.80	38.48	0.97	2.65±0.16	110±7	
Bologa	40	mcp-n	36.96	36.67	0.92	2.44±0.26	102±11	Volcanic rocks
		mcp-7	40.09	39.77	0.99	2.74±0.18	114±8	
M.-Ciuc	20	mcp-n	19.12	18.83	0.94	2.51±0.28	105±12	Sediments
		mcp-7	20.51	20.19	1	2.78±0.15	116±6	
Predeal	23	mcp-n	19.96	19.67	0.85	2.28±0.24	95±10	Sedimentary rocks
		mcp-7	22.60	22.28	0.97	2.67±0.16	111±7	
Belis	40	mcp-n	48.23	47.94	1.2	3.20±0.39	133±16	Magmatic rocks
		mcp-7	55.48	55.16	1.4	3.80±0.20	158±8	
Sinaia	23	mcp-n	18.20	17.91	0.78	2.08±0.24	87±10	Sedimentary rocks
		mcp-7	20.20	19.88	0.87	2.38±0.13	99±5	

Our relatively short term experiments, conducted for an exposure period of about three weeks have shown that this exposure time is sufficient for a clear identification of locations where the environmental dose rates are above the average value due to the natural geological substrate. Because of the low cost of the individual dosimeters, our system will allow measuring environmental gamma dose rates simultaneously at a large number of locations. Thus, it can be concluded that the successful testing of the investigated method will be a basis for obtaining realistic input data for the calculation of the effective dose received by the population in Romania. This is of paramount importance as carrying systematic surveys at a national level regarding natural exposure of the population to ionising radiation is one of the obligations of our country as a European Union member state.

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