# THE VERTICAL DISTRIBUTION OF ROCK SALT THERMOLUMINESCENCE IN THE SLANIC-PRAHOVA (ROMANIA) HALITE DEPOSITS

CLAUDIA MARIANA GOMOIU[1,a], R.M. MARGINEANU[1],
ANA-MARIA BLEBEA-APOSTU[1], B. MITRICA[1], A. JIPA[2], O.G. DULIU[2,*]

[1]Horia Hulubei National Institute of Research and Development for Physics and Nuclear Engineering,
P.O. Box MG-06, 077125 Magurele (Ilfov), Romania
*E-mail*[a]: cgomoiu@ifin.nipne.ro

[2]University of Bucharest, Department of Structure of Matter, Earth and Atmospheric Physics and Astrophysics, P.O. Box MG-11, 077125 Magurele (Ilfov), Romania
*E-mail*[*]: duliu@b.astral.ro

*Received March 6, 2014*

TL measurements, independently performed on 114 rock salt samples collected at a depth of between 130 and 210 m from the Cantacuzino mine located in the Slanic-Prahova halite deposits and on Fimel$^{TM}$ GR200A TL dosimeters, showed a monotonous decrease of the dose rate with depth in good correlation with the muon flux. The latter was calculated on the bases of experimental determinations on the salt deposits surface as well as at 210 m depth. Concurrently, the gamma spectra of salt samples collected from the same locations showed a quasi-absence of the natural radioactive elements. This, which in correlation with TL data and muon flux vertical profile points towards muon as the main depositor of energy in salt, makes the Slanic-Prahova rock-sat deposits a *sui generis* natural muon detector.

*Key words*: TL, muons, salt, dose rate, gamma spectrometry.

*PACS*: 78.60.Kn, 87.57.uq, 95.85.Ry.

## 1. INTRODUCTION

*Thermoluminescence* (TL) is a well known phenomenon consisting of a thermally stimulated light emission by some insulating crystalline materials with impurities which previously absorbed energy, mainly from ionizing radiation. As the amount of emitted light is, within some limits, proportional with the absorbed dose, TL becomes one of the most widespread methods of retrospective dosimetry [1–3]. In this regard, natural sodium chloride (the mineral halite) showed to be very suitable for TL retrospective dosimetry [4, 5].

Romania is a country rich in salt deposits, a majority of them being located in the sub-Carpathian region, when the retreat of the Sarmatian Sea determined the fractional crystallization and deposition of salt. The same tectonic processes removed the supernatant marine brine rich in potassium chloride, as well as other higher soluble

[*]Corresponding Author, fax: +4021 457 4521

salts [6]. This is the case of Slanic-Prahova halite deposits, from which 98 - 99 % purity salt [7, 8] is exploited, and whose age, according to Har *et al.* [9], corresponds to the Middle Badenian stage (cca 13 Ma). The Cantcuzino salt mine, whose exploitation begun about 22 years ago, lies within the Slanic-Prahova salt massive at a depth up to 210 m beneath the surface. The depth, as well as the extreme low content of natural radioactive elements in Slanic Prahova salt massif determined an ambient dose rate of 1.37 $\pm$ 0.3 nSvh$^{-1}$ [10], about 68 time lower than the 93.6 nSvh$^{-1}$, which is the average external dose rate recorded for Romania [11]. These circumstances make the Slanic-Prahova a potential dosimeter for the cosmic muons, the only ones which could penetrate the salt deposits.

Hence, by taking into account the presumed age of the salt massive, a simple calculus gives for the paleodose an average value of 166 Gy, way under for the saturation one would expect. Thus the vertical profile of TL signal should decrease with the depth, by following the characteristic of muons attenuation law [12]:

$$I(d) = A\left(\frac{d_0}{d}\right)^{2.5} e^{-\frac{d}{d_0}} \tag{1}$$

where: *I(d)* stays for the muon flux at depth *d* (in meter water equivalent), *A* = 0.03 m $^{-2}$s$^{-1}$ is an overall normalization constant; $d_0$ = 1470 m represents an effective attenuation length for high-energy muons [12].

This enables us to investigate to what extent the salt could be used as a dosimeter for the paleodose by measuring the depth distribution of the salts natural TL signal in correlation with the TL signals as determined by a GR220A dosimeter.

The results of our investigations will be presented further in this paper.

## 2. MATERIALS AND METHODS

### 2.1. THE SLANIC-PRAHOVA ROCK-SALT DEPOSITS

The Slanic-Prahova salt deposits consist of a lens-like accumulation whose maximum thickness (about 500 m) is reached in its central part. Due to the lateral pressure applied by the neighbouring Homorâciu anticline, a small diapir penetrating overlaying deposits appears on surface by forming different picturesque formations. Since it has been mined for centuries, the Slanic-Prahova salt deposits are now roamed by a network of galleries. This galleries facilitate access for measuring the TL signal at five different locations whose depth varied between 130 and 210 m beneath soil surface, all of them within the newly opened Cantacuzino salt mine.

We have chosen this mine for its two main advantages: i. - it consists of small, vertically aligned galleries, separated by thick ceilings so it could be considered that their volume are negligible with respect to entire salt deposit and do not significantly

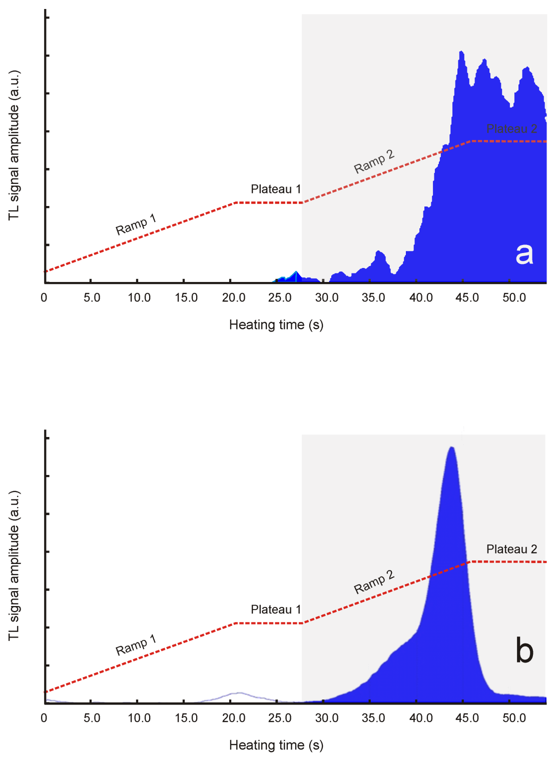


Fig. 1 – The TL spectrum of the Cantacuzino mine salt (a) and those of the GR200A TL dosimeters (b). The heating sequence is represented by lines. For both types of materials, the intensity of measured TL signal was that corresponding to the second ramp and the second plateau together and marked in darker hue on both graphs.

affects the muon flux distribution, ii. - it reaches the highest depth into the salt massif, about 210 m, so we could follow the muon flux distribution at greater depth.

Under these circumstances it was relatively easy to collect from each locations about 700 g of salt samples for further TL measurements and gamma spectrometric assays. For about 64 days, we also inserted into the gallery walls an equal number of sets of Fimel$^{TM}$GR200A TL dosimeters for comparison, each set containing six individual GR200A TL dosimeters, all of them sealed in black plastic boxes.

### 2.2. TL MEASUREMENTS

All TL measurements were performed by using a Fimel$^{TM}$TL Reader dosimetry system for environmental monitoring provided with an OEM Windows$^{®}$ LTM V.2.00 Software. In the case of rock salt we have chosen 114 individual crystals whose weights, measured by digital balance, were between 0.04 and 0.06 g.

To acquire the salt TL spectrum, each aliquot was heated by using the same sequence as those recommended for the GR200A type TL dosimeter, *i.e.* 20 s preheating at a constant rate of 6 $^{o}$C s$^{-1}$ followed by a 7 s plateau at 140 $^{o}$C continued by another heating with the same rate for 17 s and finally kept for 10 s at 245 $^{o}$C. Both heating procedure, as well as a typical TL spectrum obtained in these conditions are illustrated in Fig. 1. Moreover, we should mention that the intrinsic TL signal was that recorded for 27 s, *i.e.* between 140 and 245 $^{o}$C during the second ramp and the second plateau.

### 2.3. GAMMA RAY SPECTROMETRY

To obtain a maximum signal-to-noise ratio in recording the gamma spectra of the natural rock salt, all spectrometric measurements were performed in the Slanic Low-Background Radiation Laboratory [13]. In all cases we have used a 500 cm$^3$ Marinelli beaker filled with 600 g of material for a counting time of 5 10$^5$ seconds. All other details regarding the gamma spectrometric chain can be found in ref. [14].

### 2.4. MUON FLUX SIMULATION

To check our hypothesis that the entire Slanic salt deposit acts like a muon dosimeter, we had to calculate the depth distribution of muon flux in order to compare it with TL data of both salt samples and GR200A dosimeters (Table 1).

Previous measurements performed on surface and in the Cantacuzino salt mine at a depth of 210 m (about 1000 $\pm$ 11 m water equivalent) gave for the muon flux a value of 142.7 $\pm$ 4.9 and 0.11 $\pm$ 0.02 m$^{-2}$ s$^{-1}$ respectively [15]. This data allowed us to calculate, by means of equation (1), the vertical distribution of muon flux. As muon flux also varies with altitude, in calculating its values, we have taken into account the altitude of each location (Table 1).

## 3. RESULTS AND DISCUSSION

The experimental TL spectrum of the Cantacuzino mine salt samples are illustrated in Fig. 1a. As it is apparent, the spectrum presents more maxima with respect to LiF GR200A TL dosimeter (Fig. 1b), which shows a different spectrum, this fact being explainable by the completely different conditions under which TL materials were formed. The professional GR200A TL dosimeter consists of a high purity lithium fluoride to which controlled amounts of Mg and Ti were added, while the natural natrium chloride, although of 98 - 99 % purity, still contain unknown amounts of other elements which could determine the existence of different impurity levels and thus more TL maxima.

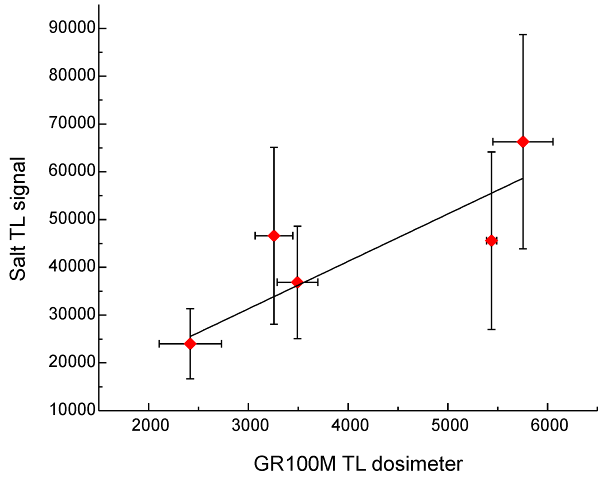


Fig. 2 – The correlation between TL signal amplitudes of GR200A dosimeter and salt.

Regardless of this fact, we have noticed a good correlation between the amplitude of TL signal produced by rock salt and GR200A dosimeters as illustrated in Fig. 2. The numerical values of TL for both rock-salt and GR200A TL dosimeter are reproduced in the Table 1. We noticed that, for all collecting point there was a monotonous decrease in the amplitude of TL signals. We have observed the same peculiarity on the vertical profile of muon flux computed as previously mentioned.

On the other hand, the gamma spectrometric data regarding the salt content of natural radioactive elements indicates for almost all samples a quasi-absence of natural radioactive elements (Table 2).

Moreover, the matrix of the correlation coefficients regarding the vertical pro-

*Table 1*

The vertical profile of the numerical values of salt and GR200A dosimeter TL signals as experimentally measured in the Cantacuzino salt mine. The fourth column contains the corresponding values of muon flux calculated by using the Eq. 1 and the experimental value presented in Ref. [15]. In the last case, the uncertainties were less then 1.5 %. Depth expressed in m, altitude expressed in meters above sea level and muon flux expressed in $m^{-2}$ $s^{-1}$.

| deph | altitude | GR200A | salt | muon flux |
|---|---|---|---|---|
| 130 | 267 | $5753 \pm 301$ | $66287 \pm 22416$ | 0.30 |
| 146 | 251 | $5438 \pm 51$ | $45565 \pm 18575$ | 0.24 |
| 162 | 235 | $3257 \pm 188$ | $46608 \pm 18499$ | 0.21 |
| 178 | 210 | $3492 \pm 204$ | $36844 \pm 11770$ | 0.16 |
| 210 | 187 | $2418 \pm 312$ | $24013 \pm 7324$ | 0.11 |

*Table 2*

The activity concentration (in Bq $kg^{-1}$) of the main natural radionuclides in salt samples whose TL spectrum amplitudes are reproduced in Table 1. The detection limits for $^{226}$Ra and $^{228}$Ac were of 0.20 and 0.05 Bq $kg^{-1}$, respectively.

| deph (m) | $^{40}$K | $^{226}$Ra | $^{228}$Ac |
|---|---|---|---|
| 130 | 6.54 ± 0.32 | 0.81 ± 0.13 | 0.30 ± 0.02 |
| 146 | 0.65 ± 0.06 | bdl | bdl |
| 162 | 0.52 ± 0.06 | bdl | bdl |
| 178 | 0.91 ± 0.09 | bdl | bdl |
| 210 | 0.74 ± 0.08 | bdl | bdl |

files data reproduced in Table 1, points toward a good correlation between all three profiles, all correlations being significant at $p \leq 0.01$ to 0.08 (see Table 3). This fact supports our hypothesis that the observed TL signal of rock salts is actually due to the natural radioactive elements, as well as to muons whose attenuation in salt deposit positively correlates with the TL signal of both salt and GR200A probes.

In the Cantacuzino mine we have noticed that both salt and GR200A TL signals decrease from - 130 m horizon to -210 m horizon by about three times, approximately the same value with the muon flux, fact which could be interpreted as proof that the muons contribution to the absorbed salt dose.

Consequently, the Slanic-Prahova salt deposits, in view of its high purity, could be regarded as a huge TL dosimeter where both cosmic rays through secondary muons generated in the the upper atmosphere together with the natural radioactive elements contributed to the absorbed dose.

## 4. CONCLUDING REMARKS

The Slanic-Prahova rock salt (halite) deposits, significantly depleted in major natural radioactive elements presents a dose rate as experimentally determined about

*Table 3*

The matrix of the Pearson's *p* correlation coefficients regarding the vertical distribution of rock salt TL signal, GR200A dosimeter TL signal as well as muons flux.

| TL dosimetric media | NaCl | GR200A dosimeter |
|---|---|---|
| GR200A | 0.8318 (0.08) | - |
| Muon flux | 0.9755 (0.01) | 0.9122 (0.03) |

The numerical values of the *p* parameter for which the correlations are significant are reproduced between brackets

68 times lower than at the surface. Under these circumstances, the salt deposits could be considered as an appropriate dosimeter for cosmic ray muons.

This hypothesis was checked by measuring at different depth of the salt massif the salt TL signal in correlation with the local dose rate as determined by using professional of GR200A TL dosimeters.

The final result of these measurements proved that the amplitude of both TL signals monotonously decreases with the depth, and at the same time, positively correlates with the computed muon flux values. Moreover, the gamma spectrometric determinations evidenced an almost negligible content of natural radioactive elements.

The observed results concerning the quasi-absence of natural radioactive elements as well as the monotonous decrease with depth of both salt and professional dosimeter TL signal were in good correlation with the calculated values of muon flux. This phenomenon could be further explained by considering that the energy deposited in salt also comes from atmospheric muons, the Slanic-Prahova halite deposit acting as a natural, *sui generis*, cosmic ray dosimeter.

*Acknowledgements.* This work was supported by the Sectoral Operational Programme Human Resources Development (SOP HRD), financed from the European Social Fund and by the Romanian Government under the contract number SOPHRD/107/1.5/S/82514 as well as by the European Regional Development Funds and co-financed by the Government of Romania - Ministry of Regional Development and Public Administration, in the framework of project ”EMERSYS Toward an integrated, joint cross-border detection system and harmonized rapid responses procedures to chemical, biological, radiological and nuclear emergencies”, MIS-ETC code 774. We would like to thank the National Society of Salt S.A (Salrom) for support in sample collecting. One author, C.M.G. wishes to acknowledge that this work was perform in fulfilment of her Ph.D. studies with the Doctoral School on Physics of the University of Bucharest. Also we would like to thank Mariana Marinescu for her help in editing the manuscript.

## REFERENCES

1. M. Aitken, *Thermoluminescence Dating* (Academic Press, 359 pp, ISBN: 978-0120463817, 1985).
2. C. Furreta, *Handbook of Thermoluminescence* (World Scientific, 480 pp, ISBN: 978-9812382405, 2003).
3. E.A. Ainsbury, E. Bakhanova, J.F. Barquinero *et al.*, Rad. Prot. Dosim. **147**, 573-592 (2011) DOI:10.1093/rpd/ncq499.
4. Y. Rodriguez-Lazcano, V. Correcher, J. Garcia-Guinea, Luminescence emission of natural NaCl, Rad. Phys.Chem. **81**, 126-130 (2012) DOI:10.1016/j.radphyschem.2011.07.012.
5. D. Mesterházy, M. Osvay, A. Kovács, A. Kelemen, Rad. Phys. Chem. **81**, 15251527 (2008) DOI:10.1016/j.radphyschem.2011.11.051.
6. C. Neniţescu, *General Chemistry* (in Romanian, Didactic and Pedagogic Press, Bucharest, 1972).
7. SNS Saltrom, Internal analysis bulletin no. 145B, (2006).
8. C. Cristache, C.A. Simion, R.M Margineanu, O.A. Culicov, M.V. Frontasyeva, M. Matei, O.G. Duliu, Radiochim. Acta **97**, 333337 (2008) DOI:10.1524/ract.2009.1622.

9. N. Har, O. Barbu, V. Codrea, I. Petrescu, Studia Univ. Babes-Bolyai (Geologia) **51**, 29-33 (2006).
10. R.M. Margineanu, A.M. Apostu, O.G. Duliu, S. Bercea, C.M. Gomoiu, C.I. Cristache, Appl. Rad. Isot. **67**, 759761 (2009) DOI:10.1016/j.apradiso.2009.01.030.
11. UNSCEAR Sources and effects of ionizing radiation, *The UNSCEAR 2000 Report to the General Assembly*, **vol I**, United Nations Scientific Committee on the Effects of Atomic Radiation, Vienna, Austria (2000).
12. J.A. Formaggio, C.J. Martoff, Rev. Nucl. Part. Sci. **54**, 361-412 (2004) DOI:10.1146/annurev.nucl.54.070103.181248.
13. R. Margineanu, C. Simion, S. Bercea, O.G. Duliu, D. Gheorghiu, A. Stochioiu, M. Matei, Appl. Rad. Isot. **66**, 1501-6 (2008) DOI:10.1016/j.apradiso.2008.04.002.
14. R.M. Margineanu, O.G. Duliu, A-M. Blebea-Apostu, C. Gomoiu, S. Bercea, Environmental dose rate distribution along the Romanian Black Sea Shore, J. Radioanal. Nucl. Chem. DOI:10.1007/s10967-013-2545-4.
15. B. Mitrica, D. Stanca, M. Petcu *et al.*, Adv. High Energ. Phys. **2013**, Article-ID 256230, 7 pp (2013) DOI:10.1155/2013/256230.