MEASUREMENTS OF RADON AND THORON DECAY PRODUCTS IN AIR.
AN APPLICATION OF LSC AND TLD METHODS*

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Liquid Scintillation Counting (LSC) is a measuring technique, broadly applied in environmental monitoring of different radionuclides. One of the possible applications of LSC is the measurement of radon and thoron progeny. There are certain advantages of this method, especially high counting efficiency for alpha and beta particles emitted by radon and thoron progeny. This advantage has been pointed out several years ago, when such methods were applied to calibration of portable monitors for radon progeny, especially due to the fact that in the case of radon progeny no standard atmosphere exists. Radon and thoron progeny might be collected on a filter, which after immersion in the liquid scintillator becomes transparent and can be counted without significant quenching. Therefore such a method can be stated as an absolute one and can be widely used for radon progeny monitoring.

Both methods, LSC and TLD, can be used for calibration of decay product monitors. The LSC method has the advantage to be an absolute one, the TLD method to measure directly the (dose relevant) deposited energy.

Key words: radon progeny, thoron progeny, LSC, TLD.

1. INTRODUCTION

The radiation risk caused by radon (radon progeny) was primarily recognized in uranium mines. Therefore at first, methods of measurement of radon progeny concentration and instrumentation were developed for uranium industry [1, 2]. Later, elevated radon and radon daughters’ concentrations were found in other types of underground mines. For instance in Poland monitoring of radon in ore mines have been started in late 70’s by means of passive track detectors and portable monitors RGR. In the beginning of 80’s a research programme, concerning radon and radon progeny in coal mines have been started by Laboratory of Radiometry in the Central Mining Institute [3]. Due to special requirements of


instrumentation intrinsic safety in Polish coal mines a special integrating monitor of PAEC was developed [4]. In this device, called ALFA-31 sampling probe, thermoluminescent detectors (CaSO₄:Dy) were applied for alpha particles counting. Three independent heads are placed over the membrane filter (FM-1) in the dust sampler’s microcyclone. Such solution enables simultaneous measurements of PAEC and dust content. Moreover, the information which is stored in TLD chips, this is the energy of alpha particles not the number of counted particles. Therefore the readout of TL detector shows directly potential alpha energy, with no dependence on equilibrium factor etc. Moreover, the implementing of that device into coal mines was approved by mining authority in Poland.

The problem is often the calibration of radon decay products monitors, while in case of thoron progeny it is even a bigger problem, a lack of primary standards of radon progeny or thoron progeny is leading to the conclusion that the best calibration method should be an absolute one. Therefore our efforts have been focused on liquid scintillation technique, in which measurements of alpha and beta particles with efficiency close to 100% are possible. The biggest problem it could be a quenching effect of a filter in the scintillator, but a membrane filter become transparent in a toluene based liquid scintillator [5]. Such method has been applied at first as the calibration method only, because at that time there were no portable LS counters.

The basic of the method is described elsewhere. Using appropriate formulas and results of measurement of the filter in liquid scintillation counter is enabling calculations of radon and thoron progeny concentrations as well as potential alpha energy concentrations.

The following approach is proposed for application of LSC for radon and thoron measurements. For radon progeny measurements, the best method seems to be the Thomas algorithm [6]. It requires three consecutive measurements done after sampling. The first period is between 2 and 5 minutes, the second one between 6 and 20 and the last one between 21 and 30 minutes after sampling. Two additional counting periods must be added to allow calculations of the two relevant thoron progeny also. Out of these two periods, first (fourth) counting period should be established rather quickly after the periods mentioned earlier to measure sampled ²¹²Bi, but should last relatively long (for instance 30 minutes). The last counting must be performed for a longer time also (30-60 minutes) but after the decay of radon progeny in the sample and with ²¹²Bi being in equilibrium with ²¹²Pb. Therefore the fifth period can be set for instance as 240 to 300 minutes after sampling.

For long term measurements a different technique can be applied – monitors of Potential Alpha Energy Concentration (PAEC) with Thermo Luminescent Detectors (TLD). In these devices, called ALFA-2000 sampling probe, TL detectors (CaSO₄:Dy) are applied for alpha particles counting. Three independent heads are placed over the membrane filter in a dust sampler’s microcyclone. Such
solution enables simultaneous measurements of PAEC and dust content. Moreover, the information which is stored in TLD chips is the energy of alpha particles, not the number of counted particles. Therefore the readout of TL detector shows directly potential alpha energy, with no dependence on equilibrium factor etc. This technique, which had been used only for radon progeny measurements, was modified to allow simultaneous measurements of radon and thoron PAEC.

1.1. APPLICATION OF THE LS COUNTER FOR RADON PROGENY MONITORING IN AIR

One of the possible applications of liquid scintillation counting is measurement of radon progeny. There are certain advantages of this method, especially high counting efficiency for alpha and beta particles, emitted by $^{218}\text{Po}$, $^{214}\text{Pb}$, $^{214}\text{Bi}$ and $^{214}\text{Po}$. This advantage has been pointed out several years ago, when such method has been applied as a calibration method of portable monitors of radon progeny [5, 7], especially due to the fact that in case of radon progeny exists no standard atmosphere. On the other hand, alpha and beta particles are counted in liquid scintillator with the efficiency close to 100 percent. Radon progeny might be collected on the filter; filter after immersion in the liquid scintillator became transparent and could be counted without significant quenching. Therefore such method can be stated as the absolute one and is suiting perfectly for radon progeny monitoring. Since 1985 [5] this method has been applied for calibration of portable radon progeny monitors as well as for calibration of TLD devices, designed and used for radon progeny monitoring in underground mines in Poland [4]. For the field measurements like in dwellings, at open air (outdoor) etc., the main drawback for a long time was the lack of the portable LS counters to allow such measurements. It is difficult to use LSC method in mines, due to the problem with methane hazard.

In last few years new instruments appeared, portable ones, therefore the monitoring of radon progeny in different environments is possible. One of the portable monitors is Triathler, produced by Hidex Company (Finland). This LS spectrometer has been used in our investigations as well as another LS spectrometer Quantulus, produced by PerkinElmer.

1.2. MEASUREMENT OF RADON AND THORON PROGENY BY LSC METHODS

The method of simultaneous measurement of radon and thoron progeny in air by means of LSC technique can be described as follows:

- For the sampling of air for radon and thoron progeny a pump with high flow-rate is necessary (at least 30–40 l/min.) and filters, which become transparent in toluene based liquid scintillator (membrane or fibreglass
4 Measurements of radon and thoron decay products in air

filters). When the calibration is done in the chamber with higher concentration of radionuclides, a smaller pump can be used to avoid significant changes of the equilibrium level during sampling.

The sampling of aerosols is done during 10 minutes and after that filter with collected aerosols is merge into vial with liquid scintillator and measured. Five consecutive measurements should be done:

- 2–5 minutes after sampling (mostly for $^{218}$Po) – $N^1$
- 6–20 minutes after sampling – $N^2$
- 21–30 minutes after sampling – $N^3$
- 40–70 minutes after sampling – $N^4$
- 240–300 minutes after sampling (mostly for $^{212}$Pb) – $N^5$

As a result five different numbers of counts are obtained, on this basis the calculation of the concentration of particular nuclides can be done as well as the estimation of potential alpha energy concentrations. The formula is following:

\[
\begin{bmatrix}
C_A \\
C_B \\
C_C \\
C_{aRn} \\
C_{aTn}
\end{bmatrix} =
\begin{bmatrix}
5.6942 & -3.1437 & 3.8919 & -0.2893 & 0.0108 \\
0.0856 & -0.7215 & 2.1662 & -0.3861 & 0.0369 \\
0.0954 & -0.0398 & 0.5977 & -0.2450 & 0.0437 \\
3.81 \cdot 10^{-3} & -4.05 \cdot 10^{-3} & 9.90 \cdot 10^{-3} & -1.83 \cdot 10^{-3} & 0.41 \cdot 10^{-3} \\
0.0269 & -0.0372 & 0.0942 & -0.0189 & 0.0148 \\
-0.7671 & 1.0618 & 2.6750 & 0.5267 & -0.0727 \\
-3.02 \cdot 10^{-3} & 4.07 \cdot 10^{-3} & -1.05 \cdot 10^{-3} & 2.04 \cdot 10^{-3} & 1.11 \cdot 10^{-3}
\end{bmatrix}
\begin{bmatrix}
N_1 \\
N_2 \\
N_3 \\
N_4 \\
N_5
\end{bmatrix} \frac{1}{V}
\]

where:

CA, CB, CC and $C_{aRn}$ – concentrations of Po–218, Pb–214, Bi–214 and potential alpha energy concentration of radon progeny;
CE, CF and $C_{aTn}$ – concentrations of Pb–212, Bi–212 and potential alpha energy concentration of thoron progeny;
V – flowrate [l/min].

1.3. RADON PROGENY MEASUREMENTS

The equilibrium factor between radon and progeny in atmosphere (especially in mines) can be very unstable and varies in space and time (we found $F$ coefficient in range 0.05–0.95). Therefore it was necessary to measure in coal mine the potential alpha energy concentration of radon progeny directly, to avoid errors due to the approximation of the exposure from radon data. For this purposes a typical, gravimetric dust sampler is used. A special sampling probe, called ALFA-31, was designed and built. The principle of the operation is as follows. During sampling aerosols are collected on the membrane filter. Thermoluminescent detectors (TLD), mounted in a special holder above the filter, measure the alpha radiation, emitted by radon progeny. The background measurements are performed by another set of
TL detectors, placed behind aluminium spacers. The sampling can be done during the whole shift and after the exposure the readout of TL detectors is performed in a special TLD reader. The calibration of ALFA-31 probes is carried out in a radon chamber with the aid of liquid scintillation method [8].

The new version of the device is called ALFA-2000 sampling probe. It can be used together with the AP-2000EX or SKC dust samplers. Therefore the applications of such devices simultaneously fulfil two tasks: measurement of dust concentration and potential alpha energy concentration (PAEC) of short-lived radon progeny. It is necessary to point out, that this technique is aimed on measurements of PAEC, but not radon gas concentration. Therefore it is possible to make comparisons only with monitors for PAEC measurements, but not with SSNTD. Participation in several intercomparison runs confirmed the precision and accuracy of the method.

Fig. 1 – AP-2000EX with a cyclone.
During measurement, the pump draws air through a filter with an adjusted flow rate. The device is equipped with a cyclone to separate out the respirable fraction and measure concentration of these particles in air.

The special sampling probe is placed inside the cyclone cassette above the filter and makes it possible to measure the PAEC value (see Fig. 2). The location of this additional part inside the cyclone does not disturb the intrinsic safety of the whole construction and measurement of dust concentration in air. TLDs are placed in these sampling probe and record radiation emitted by short-lived radon daughters bounded up with the respirable dust. The outcomes received as a result of readouts these TL detectors make possible to evaluate the PAEC of short-lived radon daughters averaged over quite long time. So this device can be used both for the environmental and personal dosimetry.

The device is supplied by a battery set and can continuously operate at least 8 hours. Measurement run is supervised by a microprocessor. There is a possibility to program the measurement time and flow rate that can be changed up to 2.3 dm³/min. The weight of this device is 550 g. The lower limit of detection related to measurement of the PAEC value is better than 0.01 µJ/m³ at a 5% significance level and 6 hours pumping. The same sampling probe was described elsewhere.

![Fig. 2 – Sampling probe Alfa-2000.](image)

Very important is the fact, that the calibration of radon progeny monitors is done with application of LSC method, but even more important is that these devices are cumulating the energy of alpha particles, emitting by nuclides, deposited on the filter. Therefore the calibration factor for the potential alpha energy concentration of radon progeny should be the same as the calibration factor for the potential alpha energy concentration of thoron progeny. It would enormously simplify the calibration of TLD monitors and measurements of PAEC values. The problem is how to separate PAECs of radon progeny and thoron progeny. It cannot be done by energy discrimination or spectrometry of alpha particles. The only solution is to make measurements with two sets of TL detectors.
The solution is based on different half-lives of radon and thoron decay products. Radon progeny is decaying faster, therefore first set of TL detectors is used just during sampling (typically 2-6 hours) and left over filter for another 4 hours afterwards. Than all TL detectors should be removed and replaced with new ones, after annealing. Another set of detects is left in the sampling probe for another 60 hours (6 half-lives of Pb-212). At the end of the process, both set are read-out in the TL reader and two PAEC values are obtained. The first one is a sum of PAEC of radon progeny and a part of thoron progeny PAEC. The second value represents another fraction of thoron progeny PAEC. Knowing the time schedule of sampling and decaying, we are able to subtract the fraction of thoron progeny PAEC from the first value and to calculate net values of radon progeny PAEC as well as PAEC of thoron progeny.

1.4. TESTING OF THE METHOD AT THE HELMHOLTZ ZENTRUM, MUNICH

During experiments in Munich we have been doing following measurements:
– measurements of PAEC in thoron chamber with application of LSC;
– measurements of PAEC with use of TLD monitor in the thoron chamber to make a cross-checking with the standard calibration, done in the Central Mining Institute, Katowice, for radon decay products PAEC;
– measurements of PAEC in the model of the clay house (traditional Chinese construction) – both LSC and TLD methods have been used for this purpose;
– measurements in some basements in the Upper Silesia region in Poland to check and verify the TLD method.

A very important problem was to find a pump with high flowrate, because the detection limit was dependent on the total amount of air, pumped through the filter during sampling. Application of fibreglass or membrane filters ensures the transparency of filters emerged in the scintillator (counting efficiency close to 100%) and enables high flow rate, dependent on the pump type and performance.

Next step was to choose a proper scintillation cocktail. In the Central Mining Institute a toluene based scintillator is used during calibration procedures. We use typical glass vials with 12 ml of the cocktail.

Than measurements of the background in wide window in Triathler and Quantulus have been done. The clean filter has been put into the scintillator and blank sample was measured. During our investigations we found rather high background of the portable instrument, ranged from 80 to 150 cpm. This is the main reason, that high volume pump must be used for air sampling for simultaneous measurements of radon and thoron progeny.

Next step was the calculation of the Lower Limit of Detection (LLD). We apply following assumptions – the flowrate is 38 l/min., counting time 10 minutes,
while the background is of about 100 cpm. The detection limit for potential alpha energy is equal LLD\(\alpha\) \(\approx\) 0.001 \(\mu\)J/m\(^3\). Despite relatively high background the detection limit is very low, because of very good counting efficiency (100%) and high flowrate, which parameters can compensate the background influence.

1.5. MEASUREMENTS OF PAEC IN THORON CHAMBER WITH APPLICATION OF LSC AND TLD

Measurements of thoron progeny with application of LSC technique and TLD devices has been made in the thoron chamber at the Helmholtz Zentrum Muenchen. Results of comparison are shown in Fig. 3.

It can be seen, that a very good agreement has been achieved, despite the fact (or due to the fact) that calibration of TLD monitors were performed in radon chamber and devices were calibrated in PAEC units for radon decay products. That comparison shows, that the cross-calibration for thoron progeny monitoring with application of TLD devices is possible and reliable.

The sampling time for TLD devices varied from 4 to 6 hours, during these periods several consecutive samplings on filters have been done, and afterwards these filters have been measured with application of LS spectrometer. In this case Quantulus LS spectrometer has been applied for measurements of filters due to the fact that concentration level of thoron progeny was relatively low. Additionally, there was no radon in the chamber and no radon decay products; therefore, it was possible to simplify the time schedule of LSC measurements.

![Thoron progeny in the thoron chamber](image)

**Fig. 3** – Results of comparison of two methods in the thoron chamber.
1.6. MEASUREMENTS OF PAEC IN THE MODEL OF THE CLAY HOUSE

The model clay house, located in the Radioecology Institute of Helmholtz Zentrum, is used for experiments, related to the monitoring and control of thoron and thoron progeny. The picture of the clay house is presented in Fig. 4a, while one of the monitoring devices is shown in Fig. 4b.

![Fig. 4 – a. Clay house; b. RAD7 monitor.](image)

**Table 1a**

Results of monitoring in the clay house

<table>
<thead>
<tr>
<th>Date and time</th>
<th>Po-218 Bq/m³</th>
<th>Pb-214 Bq/m³</th>
<th>Bi-214 Radon PAEC µJ/m³</th>
<th>Pb-212 Bi-212 Thoron PAEC µJ/m³</th>
<th>TLD Radon PAEC µJ/m³</th>
<th>TLD Thoron PAEC µJ/m³</th>
</tr>
</thead>
<tbody>
<tr>
<td>5-11-09 9:55</td>
<td>17 5 4</td>
<td>0,03 1,7 0,5</td>
<td>0,12 0,042 0,086</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5-11-09 13:25</td>
<td>17 1 1</td>
<td>0,01 1,5 2,2</td>
<td>0,12 0,029 0,099</td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>29-11-09 9:40</td>
<td>115 12 18</td>
<td>0,14 2</td>
<td>54 0,52</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>29-11-09 11:00</td>
<td>177 41 33</td>
<td>0,29 5</td>
<td>33 0,59 0,179 0,329</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>29-11-09 13:20</td>
<td>189 27 33</td>
<td>0,26 5</td>
<td>45 0,68 0,13 0,378</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Measurements were performed with application of portable LS spectrometer Triathlon, as radon and thoron progeny were present together in the clay house. Results are shown in Tables 1a and 1b.
Table 1b
Results of monitoring in the clay house

<table>
<thead>
<tr>
<th>Date and time</th>
<th>Po-218 Bq/m³</th>
<th>Pb-214 Bq/m³</th>
<th>Bi-214 Bq/m³</th>
<th>Radon PAEC µJ/m³</th>
<th>Pb-212 Bq/m³</th>
<th>Bi-212 Bq/m³</th>
<th>Thoron PAEC µJ/m³</th>
<th>TLD Radon PAEC µJ/m³</th>
<th>TLD Thoron PAEC µJ/m³</th>
</tr>
</thead>
<tbody>
<tr>
<td>30-11-09 10:20</td>
<td>163</td>
<td>44</td>
<td>20</td>
<td>0,26</td>
<td>3</td>
<td>14</td>
<td>0,29</td>
<td>0,215</td>
<td>0,236</td>
</tr>
<tr>
<td>30-11-09 12:20</td>
<td>199</td>
<td>22</td>
<td>10</td>
<td>0,20</td>
<td>3</td>
<td>37</td>
<td>0,39</td>
<td>0,180</td>
<td>0,271</td>
</tr>
<tr>
<td>30-11-09 14:10</td>
<td>165</td>
<td>30</td>
<td>26</td>
<td>0,23</td>
<td>3</td>
<td>26</td>
<td>0,37</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

It seems to be important to mention that PAEC of radon decay products was lower as PAEC of thoron decay products, while concentrations of particular decay products of radon were much higher as concentrations of thoron decay products. Moreover, values of PAEC, measured with application of TLD devices were the average values for the several hours (two parallel units have been used) while LSC values were results of grab sampling, while the sampling time was 10 minutes only.

It can be clearly seen, that results for particular nuclides are not very precise, especially in case of thoron progeny, but results of comparison of PAEC values are again very good.

1.7. MEASUREMENTS IN SOME BASEMENTS IN THE UPPER SILESIA REGION IN POLAND

To check and verify the applicability of TLD method for simultaneous measurements of radon and thoron decay products additional measurements were performed in several basements in Upper Silesia region in southern Poland. Basements with no mortar and not painted were selected for these measurements. Unfortunately, despite our efforts to find sites with higher thoron progeny concentrations, rather low concentrations of PAEC of thoron decay products have been measured, not exceeding 0,3 µJ/m³. In the same time maximum value of PAEC of radon decay products was as high as 17 µJ/m³. We are going to search for other sites, especially in areas where thorium content in the ground is much higher in comparison with the uranium content.

2. SUMMARY

Application of LSC methods for radon, thoron and radon (thoron) progeny measurements should give the opportunity to calculate equilibrium factor in dwelling, at open air, in waterworks etc. Therefore it would be a very good
possibility to assess radiation hazard in radon and thoron progeny measurements at low level, too. The method for radon and thoron progeny measurements by LSC can be used as the calibration method for other types of PAEC monitors and this can be stated as one of the biggest advantages of the method. Moreover, in case of TLD based monitors, the calibration can be done simply in radon progeny atmosphere, as thermoluminescent detectors are sensitive to energy of alpha particles.

A minor disadvantage of LSC method (especially in case of field measurements) is relatively high background of portable LS spectrometers. Therefore this effect must be compensated by use a high flowrate pump for the collection of radon (and thoron) decay products on the filters. If measurements must be done in confined places with small volume, the sampling may lead to the significant decrease of PAEC concentration, even within short time. This aspect must be taken into account during planning of experiments.

It is also possible to expand TLD method for routine measurements of thoron progeny in air. It is necessary to use longer decay time after sampling but definitely it is a very promising method, also due to simplification of the calibration procedure. It seems to be interesting to make some tests of thoron progeny measurements with application of LS spectrometer and TLD devices in the future in other laboratories, dealing with monitoring of thoron hazard.

REFERENCES