APPLICATION NOTE AN-004_EN

Thoron Measurements with the RTM1688-2

Version July 2007

Continuous and simultaneous measurement of Radon and Thoron is only possible by the measuring method of alpha spectroscopy as it is implemented in the RTM1688-2.

The collection of the short living daughter products on the surface of a semiconductor detector allows the separation of the single nuclides with respect to their emission energy.

There are still some particularities affecting the behaviour of the instrument. The knowledge of these particularities is necessary to conduct successful measurements of mixed Radon/Thoron atmospheres.

Physical Background

To perform an alpha spectroscopic analysis, the immediate Radon/Thoron daughter products Po-218 and Po-216 are ionised during the decay of the gas. The generated ions are collected on a planar semiconductor detector by an applied electrostatic field. After a short time, the activity of the daughter products on the detector is in equilibrium with the Radon/Thoron concentration within the sampled air. During a measurement, the Radon/Thoron concentration can be calculated backwards by counting the decay events of their collected daughters over a pre-set time period (determination of the activity) (see also AN-003). To separate the different daughter products from each other the method of alpha spectroscopy is used. Following nuclides can be present within in the spectrum:

From the Radon (Rn-222) decay chain:

Po-218 with an emission energy of 6.002 MeV Po-214 with an emission energy of 7.687 MeV

From the Thoron (Rn-220) decay chain:

Po-216 with an emission energy of 6.778 MeV

Po-212 with an emission energy of 8.785 MeV

Bi-212 with an emission energy of 6.050 an 6.080 MeV

The gases Radon (5.490 MeV) and Thoron (6.288 MeV) will not be visible within the spectrum because they are not collected on the detector.

An ideal spectrum would show the counts of one single nuclide as a mono-energetic line, meaning that all decay events are counted in one channel of the spectrum. However, a real spectrum gives a peak which is distributed over few channels instead of the line. The shape of the peak is similar to a Gauss function with a significant flat slope on its left side which is called "Tailing".

The Gauss shape is the result of the electronic (thermal) noise of the detector and the preamplifier. Because it is a physical property, it can be not completely suppressed. The tailing is generated by the energy lost of the alpha particles within the non-sensitive protection layer of the detector. Both, the thickness of this layer and the size of the detector determine the exact shape of the tailing. Since the daughter products are deposited on the detector surface, the distance of a particle to pass the non-sensitive zone depends on its emission angle. This results in different energy levels for various emission angles if the particle enters the sensitive zone.

To assign the registered decays, an own energy range (ROI = region of interest) for each nuclide has to be defined. The upper limit of each ROI is set a little bit higher than the emission energy of the nuclide which shall be covered by this ROI. The lower limit is equal to the upper limit of the next lower ROI which covers the nuclide with the lower emission energy. This definition ensures that the major part of the decays of the considered nuclide will be counted into the assigned ROI and that the ROI will be only affected by the tailing of the nuclides with higher energy levels.

We are faced to following situation with respect to the available nuclides:

ROI Po-212: The ROI is unaffected because it covers the nuclide with the highest possible emission energy. Po-212 is not used for the measurement of Radon and Thoron. and will be present only as a disturbance variable.

ROI Po-214: About 5% of the decays of the Po-212 are counted in addition to the Po-214 decays. High Po-212 activities resulting in a background during the measurement of Radon if the slow mode has been selected.

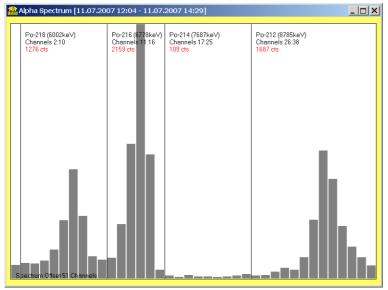
ROI Po-216: Beside Po-216 also 8% of the Po-214 and 2% of the Po-212 are covered by this ROI. Increased activities of Po-212 as well as Po-214 resulting in a background during the Thoron measurement.

Po-218: This ROI contains the Po-218 as well as the Bi-212. The emission energy of both nuclides is nearly identical (6.002 MeV and 6.050/6.089 MeV) so that it is impossible to separate them by alpha spectroscopy. Furthermore, approximately 20% of the Po-216 decays will appear in the ROI. High activities of Po-216 and Bi-212 generate a background during the Radon measurement.

The following figures show a couple of spectra which are pointing out the described circumstance.

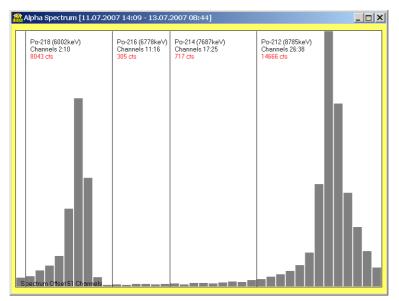
Alpha Spectrum [11.07.2007 11:39 - 11.07.2007 11:49]				
Po-218 (6002keV) Channels 2:10 393 cts	Po-216 (6778keV) Channels 11:16 1980 cts	Po-214 (7687keV) Channels 17:25 21 cts	Po-212 (8785keV) Channels 26:38 43 cts	
Spectrum Offset 51 Channels				

Thoron spectrum during the first hours of the exposure. Only Po-216 is available because Po-212 and Bi-212 will be generated with a strong delay by the half life of Pb-212 (10.64 hours). Pb-212 is beta emitter which is placed between Po-216 and Bi-212 in the decay chain. The tailing of the Po-216 within the Po-218 ROI is visible.



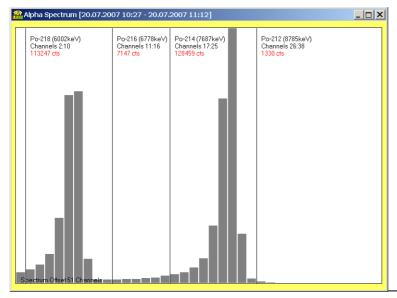
Clean Thoron spectrum in the activity equilibrium. The peaks of Po-212 and Bi-212 are fully developed. Very good to see is Bi-212 that the peak is completely covered by the Po-218 ROI. The significant slope on the right side of the Po-212 peak comes from the previous beta decay of

the Bi-212. The pulses of the alpha and the beta decay are electronically overlaid due to the short half life time of the Po-212 $(0.3 \ \mu s)$.



Spectrum after Thoron exposure. The Po-216 peak is disappeared immediately after exposure. The peaks of Po-212 and Bi-212 are still present over few days due to the Pb-212 which has been enriched during the exposure period. It decays with a half life time of 10.64 hours and generates new Po-212 and Bi-212 during this time.

The spectrum shows also the tailing of Po-212 within the Po-214 and Po-216 ROI.



Radon spectrum in the equilibrium state. The Po-214 tailing affects the Po-216 ROI. The Po-218 peak is shifted a little bit to left in comparison to the Bi-212 peak in the upper spectrum, conditional on its little bit lower emission energy.

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We can conclude that Radon and Thoron will affect each other during theirs simultaneous measurement. A suitable correction procedure has to be applied.

Correction Procedures

The tailings can be considered as detector specific device constants. During the calibration of each instrument, the percentage of decays of the interfering nuclides within the affected ROI's has to be determined.

For example, the tailing constant "Tailing (Po-212 > Po-214)" specifies the background which is generated by the Po-212 within the Po-214 ROI. If the instrument is exposed to a clean Thoron atmosphere, the constant can be determined with sufficient high statistical accuracy. In this case only the background of the Po-212 is present and not the Po-214 itself. In our example the ROI3 shall be assigned to the Po-214 and the ROI4 to the Po-212:

 $K_{212>214} = 100\% * N(ROI3) / {N(ROI3 + N(ROI4))}$

N(ROI3) and N(ROI4) are the detected decays of the Po-212 within the both ROI's. $K_{212>214}$ is the tailing constant "Tailing (Po-212 > Po-214)".

If a real measurement of a mixed atmosphere is conducted, the ROI3 contains the decays of the Po-214 as well as a part of the Po-212 decays. To separate the Po-214, the Po-212 decays have to be subtracted using the information about the Po-212 activity from the unaffected ROI4:

 $N(Po-214) = N(ROI3) - \{K_{212>214} / (1 - K_{212>214}) * N(ROI4)\}$

N(Po-214) means the number of decays within the ROI3 which can be assigned to the Po-216.

This procedure is applied to all ROI's.

A different physical relation has to be used in case of the Bi-214 inside the Po-218 ROI. It is impossible to create separate ROI's for both nuclides. However, the ratio between the activities of Po-212 and Bi-212 is always constant. This results from the two different decay branches of the Bi-212. While approximately 36% of the Bi-212 are converted directly by an alpha decay, the remaining 64% are converted to Po-212 by a beta decay. The generated Po-212 decays immediately with a half life time of 0.298 µs by emission of alpha radiation. Therefore, the equilibrium state between Po-212 and Bi-212 is given at any time. Then, the Bi-214 decays within the Po-218 ROI can be calculated by the formula:

N(Bi-214) = 0,5625 * N(ROI4)

N(ROI4) means the Po-212 decays within the unaffected ROI4.

Statistical Considerations about the Corrections – Detection Threshold

Because the decay process of a nuclide undergoes statistical fluctuations (see AN-003), also the background which is generated by the nuclide within the different ROI's is a statistically distributed value. The expressions used for the corrections are only valid for the mean values if the number of detected decays becomes infinite.

In practice we are faced to a finite number of counts and therefore it is required to estimate the fluctuation range in a statistical sense. To clarify the circumstance we will come back to the previous example once again.

If we presume that no Po-214 is available on the detector, the number of detected Po-212 counts within the Po-214 ROI would be different for each sample interval even if the activity remains stable. Using the correction procedure results in an either positive or negative number of Po-214 decays with a probability of 50% for each case. Since negative number of decays is impossible we would correctly suppose that there is no Po-214 on the detector. The problem are the positive results. 50% of sample intervals will show a Po-214 contribution which is not present.

To avoid such false positives, the possible fluctuation of the background has to be calculated by the rules of the statistical error propagation. We accept that Po-214 is available only if the number of decays within the Po-214 ROI is equal or higher than the estimated fluctuation range. All other positive results could contain Po-214, but it is not sure.

The statistical calculation of the fluctuation range is based on the specification of a confidence interval. The confidence interval defines the probability that the number of Po-212 decays within the ROI3 not exceeds the calculated value. A confidence interval of 95% means that in 95% of all sample intervals the detected background is less or equal to the fluctuation limit while 5% of all sample intervals will give a higher number of Po-212 decays within the ROI3. That is important: In 5% of all cases we still get a false positive result even if the correction procedure has been applied. Of course it is possible to enlarge the confidence interval. However, the probability to detect a true Po-214 contribution decreases likewise.

The smallest value which gives a confidential contribution of the wanted signal (background fluctuation limit, Po-214 in our example) is called "Detection Threshold".

Meaning of the Detection Threshold for the Measurement

Three different situations shall be considered:

Case 1: A clean Radon atmosphere, Po-212/Bi-212 are not present on the detector

The ROI's of Po-218 and Po-214 are not affected. Each detected decay can be uniquely assigned to these nuclides. The detection threshold for the fast mode Radon calculation is just one decay event within the Po-218 ROI. The detection threshold for slow mode Radon calculation is just one decay within one of the both ROI's for Po-218 and Po-214.

The Po-216 ROI is affected by the tailing of the Po-214 which is in activity equilibrium with Po-218 after approximately 3 hours. The detection limit for Po-216 increases with an increasing Po-214 activity because the fluctuation limit for the Po-214 background within the Po-216 ROI increases likewise.

The specified confidence interval of 95% leads to a not really present Po-216 contribution in 5% of all sample intervals.

As a result we have an increasing detection limit for the Thoron measurement because the Thoron measurement is based on the Po-216 decay rate. Around five of hundred sample intervals will show a Thoron concentration without any Thoron in the measured air.

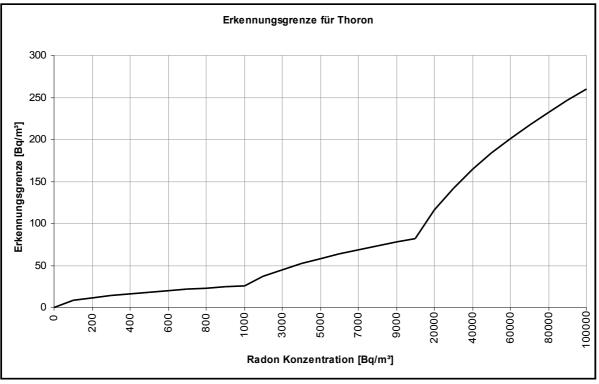


Fig. 1 Detection Threshold (2σ) for Thoron (RTM1688-2, 60 Minutes Sample Interval)

Case 2: A clean Thoron atmosphere

At the begin of the measurement only Po-216 appears within the spectrum. Po-212 and Bi-212 are not yet present due to the intermediate Pb-212 with its half life time of 10.64 hours. The Po-218 ROI is affected by the tailing of the Po-216. Similar to case 1 increases the detection threshold for Po-218 with an increasing number of Po-216 decays.

After a while, the slowly growing Po-212 activity results in a background within the ROI's for Po-214 and Po-216. Therefore, the detection limits for Po-214 and Po-216 are increasing with the growing Po-212 activity.

However, the major problem is the Bi-212 because all its decays are counted within the Po-218 ROI. That means, the detection threshold of the Po-218 increases much stronger compared to Po-214 and Po-216.

We can conclude: There is only a little bit increased detection limit for Thoron due to the interference of the Po-212 tailing. The detection threshold for Radon is strongly increased if the fast mode calculation (based on Po-218 only) has been chosen. A similar situation is given for the "slow-mode" Radon where Po-218 delivers about 50% of the signal. In this case is the exclusive usage of Po-214 the better choice. The detection threshold is much lower because only the Po-212 tailing affects this ROI. The RTM1688-2 switches over automatically if the Bi-212 background becomes too high.

Due to the estimation of the background we will get in 5 of 100 sample intervals a Radon concentration above zero even if only Radon is present.

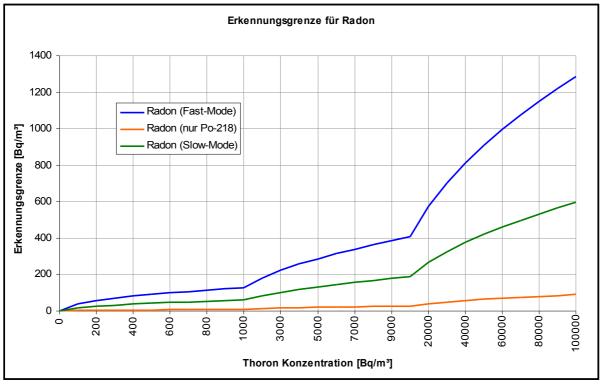


Fig. 2 Detection Threshold (2o) for Radon (RTM1688-2, 60 Minutes Sample Interval)

Case 3: Mixed atmosphere (Radon and Thoron are present)

A basic presumption for the simultaneous measurement is that the concentrations of both components are higher than their detection thresholds. The expected statistical errors of the measurement depend on the ratio of the concentrations at the one hand and the total number of counted decays within the different ROI at the other hand. The latter is dependent on the concentration level, on the selected sampling interval and on the sensitivity of the monitor. The dependence on the mixture ratio shall be discussed by means of the following graphic (Fig. 3).

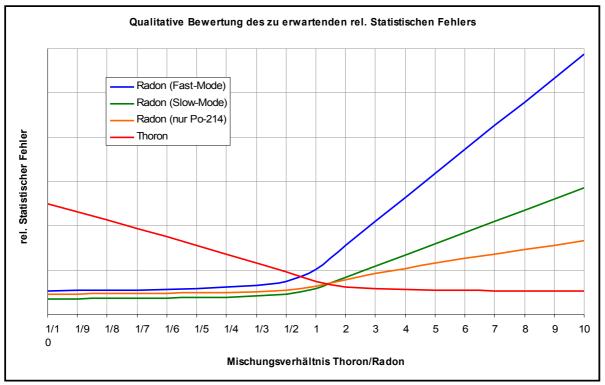


Fig. 3 Statistical Error of a mixed Atmosphere, qualitativ

The X-axis shows the mixture ratio between Thoron and Radon. A value of 4 means that the Thoron concentration is four times higher than the Radon concentration within the air while a value of 1/4 indicates a four times higher Radon concentration

The relative errors of both components are quite low for mixture ratios between $\frac{1}{2}$ and 2. The Thoron error (red line) increases if the ratio becomes lower by the increasing Radon (Po-214) background while the Radon decreases weakly. In case of ratios above 2, the Radon error increases depending on the used calculation method.

If the fast mode is used (based on Po-218 only), the error rises very soon due to the Bi-212 background and the Po-216 tailing.

Using the slow mode (green line) gives a less significant increase because the Po-214 is included in the calculation which is only slightly affected by the Po-212 tailing.

The lowest error is obtained if the Po-214 is exclusively used for the Radon calculation (orange line). In this case, the background is limited to the Po-212 tailing. Compared with the Thoron error in case of an opposite mixture ratio the increase of the orange line is smaller. The reasons are the smaller influence of the Po-212 tailing (only 5% compared with 20% for Po-214 to Po-216) and the fact that only 64% of the Po-216 will appear later as Po-212 while 36% will decay as Bi-212.

Practical use of the Monitor in case of mixed Atmospheres

The interaction between Radon and Thoron is mostly not a problem if continuous measurements are performed. If the mixture ratio exceeds the factor 100, the low concentration component is negligible from the radiation protection point of view. The only limitation is the increased response time if the slow mode radon calculation has to be used to keep the statistical error low.

The diagram in figure 4 shows the statistical error of a Thoron measurement in presence of an up to 100 times higher Radon concentration. The dependence on the actual Thoron concentration value (related to the absolute detected number of decays) is given as a function parameter.

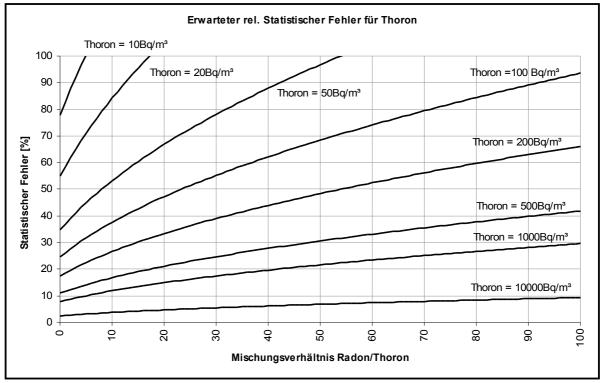


Fig. 4 Statistical Error (1o) for Thoron (RTM1688-2, 60 Minutes Sample Interval)

Example:

At a Thoron concentration of 1000 Bq/m³ and a 40 times higher Radon concentration (40000 Bq/m³), the resulting statistical error is approximately 20%.

The diagram in figure 5 shows the statistical error of a Radon measurement in presence of an up to 100 times higher Thoron concentration. The dependence on the actual Radon concentration value (related to the absolute detected number of decays) is given as a function parameter.

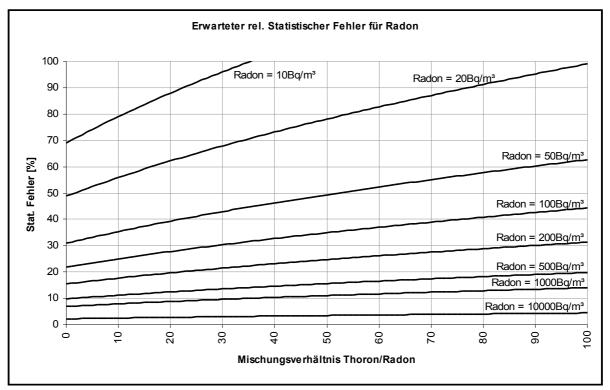


Fig. 5 Statistical Error (1o) for Radon (RTM1688-2, 60 Minutes Sample Interval)

Example:

At a Radon concentration of 200 Bq/m³ and a 20 times higher Thoron concentration (4000 Bq/m³), the resulting statistical error is approximately 20%.

Problems will arise if low Radon concentrations have to be measured after the measurement of high Thoron concentrations. After the Thoron exposure, the Pb-212 is still present on the detector and decays with a half life time of 10,64 hours. The Pb-212 daughter nuclides Bi-212 and Po-212 are generated continuously as long as the Pb-212 is available. The decay of the Po-212 and Bi-212 results in a background for the Radon measurement even if Thoron is not longer in the measurement chamber.

Therefore, please pay attention to following notes:

Start a measurement campaign at the places where the lowest concentrations are expected. Inside buildings you will commonly find the highest concentration in the seller while it decreases in the upper floors.

Carry out soil gas measurements at the end of the campaign and do it as fast as possible. Soil gas contains in the most cases high Radon concentrations and also significant amounts of Thoron.

Keep the sampling times as short as possible if Thoron is present. The measurement chamber is already flushed completely after about 5 to 6 Minutes. This time is sufficient long to measure Thoron. After that, the chamber can closed by a short hose piece between air inlet and outlet. Now, a new measurement to determine the Radon concentration of the sampled can be started.

Wait, if possible, several days after a strong Thoron exposure to conduct a new measurement campaign. During this time the Po-212/Bi-212 will decay completely.

Some sporadically appearing concentration values within a time distribution pointing in the most cases at an increased background. Then it is very likely that no real concentration is present but the result comes from the 5% false positives due to the confidence interval of the background fluctuation range. An average created from a few sample periods around the "maverick" shows mostly a value below the detection threshold.

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