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CALIBRATION OF THE POLITRACK® SYSTEM BASED ON CR39 SOLID-STATE NUCLEAR TRACK DETECTORS FOR PASSIVE INDOOR RADON CONCENTRATION MEASUREMENTS

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Swiss national requirements for measuring radon gas exposures demand a lower detection limit of 50 kBq h m^{-3}, representing the Swiss concentration average of 70 Bq m^{-3} over a 1-month period. A solid-state nuclear track detector (SSNTD) system (Politrack, Mi.am s.r.l., Italy) has been acquired to fulfil these requirements. This work was aimed at the calibration of the Politrack system with traceability to international standards and the development of a procedure to check the stability of the system. A total of 275 SSNTDs was exposed to 11 different radon exposures in the radon chamber of the Secondary Calibration Laboratory at the Paul Scherrer Institute, Switzerland. The exposures ranged from 50 to 15000 kBq h m^{-3}. For each exposure of 20 detectors, 5 SSNTDs were used to monitor possible background exposures during transport and storage. The response curve and the calibration factor of the whole system were determined using a Monte Carlo fitting procedure. A device to produce CR39 samples with a reference number of tracks using a ^{241}Am source was developed for checking the long-term stability of the Politrack system. The characteristic limits for the detection of a possible system drift were determined following ISO Standard 11929.

INTRODUCTION

Radon is a radioactive gas that is known to be the most important cause of lung cancer after smoking. Most of the radon exposure of the public takes place in closed environments at home or at work(1). To effectively manage radon risk, radon concentrations have to be estimated via reliable measurements. The most reliable radon measurements are long-term measurements that integrate between 3 and 12 months of exposure(2). There is a variety of different devices to measure radon concentrations. Solid-state nuclear track detectors (SSNTD) have proven to be cost-effective and therefore particularly suitable for large-scale national radon surveys. However, practical questions arise when putting into place an SSNTD device: how to keep track of possible drifts of the system in order to warrant a stable quality of results and how do radon concentrations during transport and storage influence the result of SSNTD readings?

INSTRUMENTATION AND METHODS

Solid-state nuclear track detectors

Heavily ionising particles leave trails of damage on most insulating materials(3). These trails can be made visible with a microscope by etching the material with a chemical reagent, which attacks preferentially the damaged trails. Detectors that work on this principle are called 'Solid State Nuclear Track Detectors (SSNTD)'. An SSNTD for alpha particles can be built with the polymer CR39 as detector material. The authors used CR39 films of a thickness of 1 mm and a size of 25 × 25 mm and etched them, according to the manufacturer recommendation, in a 6.25 M NaOH bath for 1 h at 98°C after exposure.

SSNTD reader system

After etching, the tracks of the alpha particles can be counted with a light microscope. The Politrack reader system consists of a microscope equipped with a CCD camera and a 4× magnifying objective that can be moved in z direction and an SSNTD stage that can be moved in xy direction. The images are sent to a computer via firewire and analysed by a programme written in LabView by the manufacturer. The LabView software returns the number of counted tracks per square centimeter and the sum of the area of all detected tracks. Each alpha track is detected separately by a pattern recognition algorithm. At higher exposures, the probability increases that two or more alpha tracks overlap. This leads to a saturation effect. The algorithm is not capable to distinguish between two overlapping tracks. Since the probability to obtain overlapping tracks is difficult to model, track overlapping was corrected by the empirical formula:

$$\mathrm{Tr}_{\mathrm{cor}} = \frac{\mathrm{Tr}_{\mathrm{net}}}{1 - mA_{\mathrm{Tr}}}, \tag{1}$$

where m is an empirical factor to be determined by a fit procedure, $\mathrm{Tr}_{\mathrm{net}}$ is the difference between the counted tracks and the background and A_{Tr} is the area of all detected tracks. The area was corrected by the mean area measured on the background SSNTDs.

CALIBRATION

The authors exposed 275 SSNTDs at 11 different exposures in the radon chamber at the Paul Scherrer Institute in Villigen, Switzerland[4]. The exposures took place consecutively at three different concentration levels: 1000, 5000 and 20 000 Bq m^{-3}. The different exposures within each concentration level were realised by different exposure times resulting in the exposures: 46, 96, 289, 502, 987, 1046, 2077, 3112, 5230, 10 506 and 15 520 kBq h m^{-3}, with an expanded uncertainty ($k = 2$) ranging between 1.4 and 2.3 %. After exposure, the SSNTDs were shipped back to the Institute of Radiation Physics, Lausanne, and stored until the exposure of the last SSNTDs finished. The SSNTDs were etched at three batches. In order to keep track of background exposure during transport and storage, each exposure level was accompanied by five transport SSNTDs.

Taking into account the correction of Equation (1), the following model can be fitted to the background corrected track counts $\mathrm{Tr}_{\mathrm{net}}$, A_{Tr} and the reference exposure E to obtain the empirical factors c and m, where c is the calibration factor:

$$E = c\frac{Tr_{\mathrm{net}}}{1 - mA_{Tr}}. \quad (2)$$

To take into account the uncertainty due to random errors of the exposure and the track reading, the authors used a Monte Carlo fit procedure according to (5). For this purpose, they repeated least squares fitting 10 000 times by adding each time a random error $\varepsilon_{\mathrm{Exp}}$ to each exposure value with:

$$\varepsilon_{\mathrm{Exp}} \in N(0, u_{\mathrm{Exp}}), \quad (3)$$

where u_{Exp} is the standard uncertainty estimated for the reference exposure of each SSNTD.

Furthermore, for each of the 10 000 repeated fittings, the authors added a random error ε_{Tr} to the track counts $\mathrm{Tr}_{\mathrm{net}}$ in order to account for the reading uncertainty with:

$$\varepsilon_{\mathrm{Tr}} \in N(0, u_{\mathrm{Tr}}), \quad (4)$$

where u_{Tr} depends on the number of tracks of each readout and was given by the manufacturer.

The authors took the arithmetic mean and the standard deviation of c and m over the 10 000 fitting results as best estimates for the expected values and the corresponding standard uncertainties of c and m.

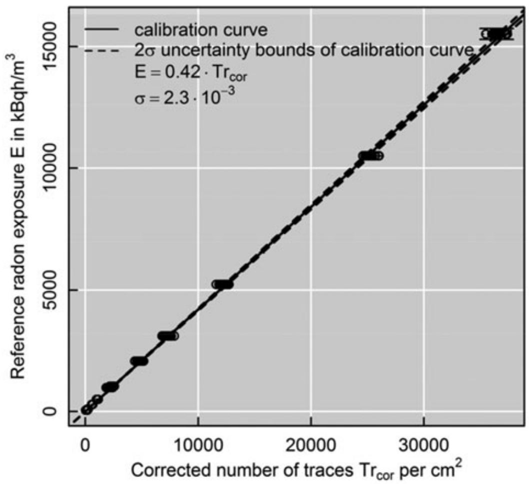

Figure 1. Calibration curve of reference exposures versus corrected tracks.

Figure 1 shows the calibration curve that the authors fitted by the Monte Carlo fit procedure. For simplicity, the authors plotted the exposure E versus $\mathrm{Tr}_{\mathrm{cor}}$. The uncertainty u_{Tr} due to read out of the system was given by the vendor for each SSNTD read out and ranged from 0.7 to 13 %. The expanded uncertainties ($k = 2$) of the reference exposures, corresponding to u_{Exp}, ranged between 1.4 and 2.3 %.

The Monte Carlo fit procedure yielded a calibration factor $c = 420.0 \times 10^{-3}$ kBq h m^{-3} cm^2 with an uncertainty of $u_c = 2.3 \times 10^{-3}$ kBq h m^{-3} cm^2 (0.6 %) and a track correction factor $m = 5.55$ cm^{-2} with a standard uncertainty of $u_m = 0.07$ cm^{-2} (2.7 %). Hence, the overall uncertainty of c an m results in 0.93 %. The uncertainty u_m contributes only to a small amount, since A_{Tr} is generally very small. 0.93 % indicates a small overall uncertainty for the calibration factor and is due to the fact that the uncertainty is attributed to random errors, which have a very small influence on the final fit result, since the authors used a relatively large number of SSNTDs for the calibration. For simplicity, they did not assume an intercept in Equation (1). The Monte Carlo fit procedure would, however, allow to calculate a covariance between the intercept and the slope of the calibration curve, which could be used to further improve the uncertainty estimation.

The authors assumed a maximum u_{Exp} of 1.15 % as systematic standard uncertainty contribution from the calibration of the radon chamber.

LONG-TERM STABILITY MONITORING

To monitor possible drifts of the system, the authors developed and built a device to produce reference CR39 films at a reproducible exposure level. For this purpose, they used a ^{241}Am source with an activity of 320 Bq. To control the exposure time of the ^{241}Am source, they shielded the source with an automatic shutter.

The distributions of track counts for several reference exposure series are presented as boxplot in Figure 2. The boxplot represents the median, first and third quartile. The whiskers represent 1.5 times the interquartile range of the distribution. For further readings on boxplots, refer to (6). Each batch has been etched separately. The authors annotated the basic statistical indicators for each batch on the boxplot. The uncertainty of the track counts is attributable to the randomness of the ^{241}Am decay, to variation in the etching procedure as well as to read out uncertainties.

The decision threshold for potential drifts was determined according to ISO 11929 on international standards[7]. The decision threshold Tr^*_{net} can be determined by the equation:

$$Tr^*_{net} = q_{1-\alpha}\,\tilde{u}, \qquad (5)$$

where $\tilde{u}$ is the standard uncertainty of the average number of tracks of the irradiated reference SSNTDs and $q_{1-\alpha} = 2.33$ the $(1-\alpha)$—quantile of the standardised normal distribution. The authors chose $\alpha = 1$ % and assumed the relative decision threshold Tr^*_{net} as uncertainty contribution resulting from long-term stability monitoring of the system. For the irradiation of 10 reference SSNTDs with the ^{241}Am source for 20 s, they observed a mean of ~780 cm^{-2} with a standard uncertainty of 14 cm^{-2}. This results in a decision threshold for a possible drift of 32 cm^{-2} and hence gives an uncertainty contribution on Tr_{net} of 4 % due to long-term stability monitoring.

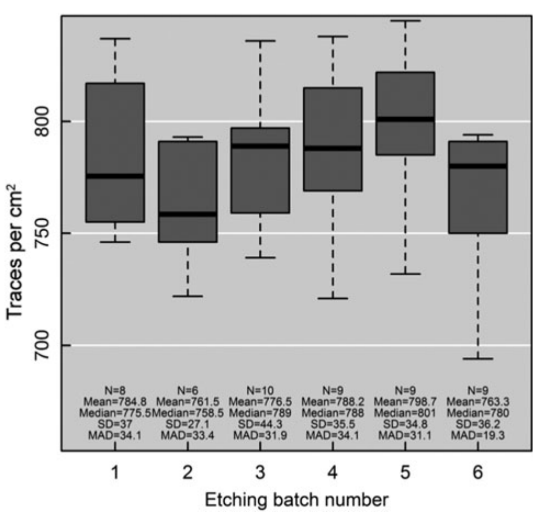

Figure 2. Track count distributions of separately etched SSNTD batches used for reference exposure series.

OVERALL UNCERTAINTY OF THE SYSTEM

Table 1 shows the uncertainty budget of the estimated exposure *E*. The summation in quadrature leads to an expanded combined uncertainty ($k = 2$) of ~9 %

By far the largest uncertainty contribution results from the long-term stability monitoring with 4 %. This is reasonable, since the etching procedure is a process that is difficult to control.

LEAKAGE OF SSNTD WRAPPINGS

To control the air tightness of the SSNTD packaging, 40 SSNTDs welded up in plastic bags were exposed to the highest exposure of 15 520 kBq h m^{-3}. Some of the weld seams showed small defects. To keep control of a possible background contribution, 10 SSNTDs welded up in plastic bags were used that were exposed simultaneously to ambient air. After exposure, the authors distinguished between SSNTDs packed in plastic bags with defects (Weld defect), with no defects (No weld defect) and background SSNTDs (Transport SSNTD) and compared their distributions via boxplots.

The results are shown in Figure 3. A Kruskal–Wallis analysis of variance comparing the medians of the three groups yielded a $\chi^2 = 1.93$ and a *p*-value of 38 %. This result does not support the hypothesis of leakage of detector wrappings. This holds for regular weld seams as well as for weld seams exhibiting little defects. Since the authors observed 41 cm^{-2} on the 10 transport SSNTDs that were exposed to ambient air in plastic bags, they assumed a general background correction of all readings of 40 cm^{-2} for the comparison measurements described in the next section.

COMPARISON MEASUREMENT

In order to compare the authors' calibration with commercially available SSNTDs, they carried out a comparison study with their SSNTDs and SSNTDs well established on the market (Landauer Nordic, former Gammadata). For this purpose, they distributed 30 SSNTDs of each type pairwise in Swiss

Table 1. Uncertainty budget of estimated exposure *E*.

Uncertainty component	Relative standard uncertainty (%)
Long-term stability monitoring	4
Reference exposure values	1.15
Monte Carlo fit procedure	0.93

schools and measured the radon concentration for ~3 months.

The comparison of measurements with Politrack SSNTDs with the authors' calibration factor and Gammadata SSNTDs shows a mean difference of 14.1 kBq h m^{-3} (Figure 4). That corresponds to a concentration of 6.5 Bq m^{-3} for a measurement of ~90 d. This is indicating a good accordance of both systems.

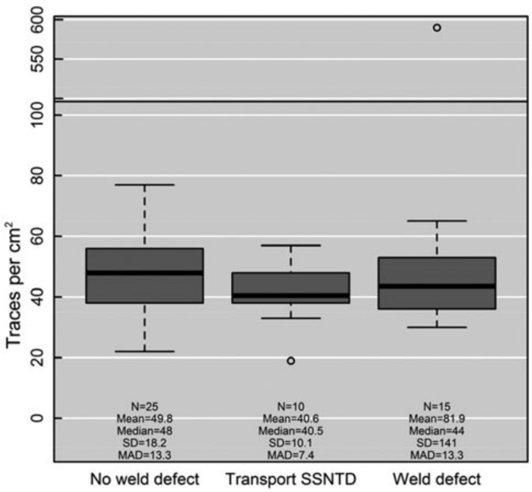

Figure 3. Boxplot of track count distributions of SSNTDs that were packed in plastic bags with and without weld seam defects. The boxes represent the median, first and third quartile. The whiskers represent 1.5 times the interquartile range of the distribution. Data points that lie outside of the whiskers are drawn on the plot as extreme values. The plastic bags with the SSNTDs were exposed to 15 520 kBq h m^{-3}. The track count distribution of the SSNTDs that were only exposed to background radon concentrations is indicated as 'Transport SSNTD'.

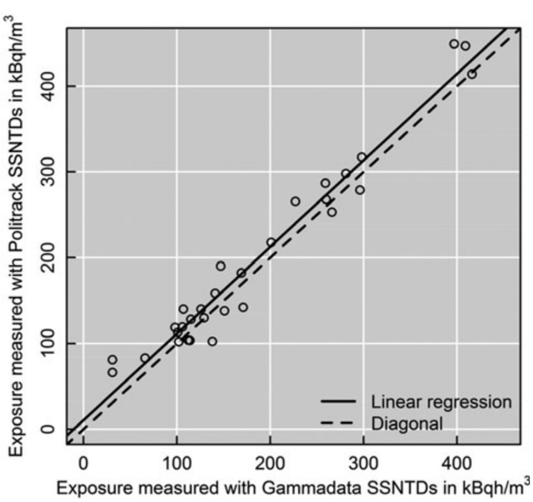

Figure 4. Comparison of Politrack with Gammadata SSNTDs.

CONCLUSION

The authors calibrated and characterised an SSNTD reader system for the measurement of indoor radon concentrations. In addition to that, they developed a procedure to keep track of long-term stability of the system. The system is ready for routine use.

The authors achieved an overall uncertainty of ~9 % for the system. A comparison in school buildings with SSNTDs of the manufacturer Landauer Nordic yielded consistent results with their calibration.

The authors' results indicate that the weld of the SSNTD transport plastic bags is sufficiently air tight for shipping. After an exposure of packaged detectors to ~15 000 kBq h m^{-3} during 31 d, the authors did not find significantly higher mean track counts than those for detectors that were only exposed to background irradiation. This also holds for plastic bags having defects in the weld seam.

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