ALPHA PARTICLE EMISSION FROM REFERENCE GLASS SURFACES IMPLANTED WITH $^{210}\text{Po}$

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Implanted long-lived radon decay products in glass surfaces have been used as a measure of past radon exposure in homes. Special track-etch devices (so-called "retro-detectors") attached to the glass surface, have the ability to specifically measure the implanted activity of $^{210}\text{Po}$ in-situ. Calibrating these devices for $^{210}\text{Po}$ is fairly straightforward, but the retro-detectors are also sensitive to the background activity of the glass substrate. Thus, for the successful calibration of retro-detectors, it is necessary to determine the complete alpha emission energy spectrum of the reference glass sheet utilized as a calibration pad. In order to achieve accurate knowledge of the alpha surface emission rate, we have combined several different approaches, i.e. alpha spectrometry of the pad surface with both surface-barrier and pulse-ionisation detectors, and activity determination of the glass matrix by means of radiochemical methods. The part of the alpha emission spectrum originating from the glass volume is then calculated theoretically and compared with experimental results.

Keywords: $^{210}\text{Po}$, Radon, Retrospective, Alpha-spectrometry, Track-etch, Pulse-ionisation chamber

INTRODUCTION

The behaviour of radon gas and its decay products in the indoor environment have been known, at least qualitatively for many years. Depending on the length scale and milieu we are interested in, the difficulty in calculating of the local concentration of radon decay products theoretically ranges from the labourious to the downright impossible. The need for a much better and more detailed understanding of radon gas and radon daughter behaviour indoors, is in fact raised by the new RARE (Retrospective Assessment of Radon Exposure) approach, which utilises the long-lived radon progenies trapped and accumulated in objects found around the home (Samuelsson, 1988). The key factor in this approach is how well the long-lived activity trapped inside (including superficially implanted) different objects, correlates with the cumulated airborne activity inhaled by the inhabitants. Even if we happen to know the occupancy factors and the inhalation patterns of the individual inhabitants, we are left with the task of converting the long-lived daughter activity of the objects measured, to the time-integrated airborne concentration of radon gas or its short-lived progenies. Track-etch detector assemblies (retro-detectors), specifically devoted to measuring $^{210}\text{Po}$ implanted into plane surfaces, are a useful tool for elucidating the long-term behaviour of short-lived radon progenies. The experimental information thus obtained is useful in radon risk estimates, as well as in radon room-model calculations. The main objective of this paper is to illustrate how these track-etch devices are calibrated in the laboratory, using a pulse-ionisation detector (PIC) specially developed for large-area alpha spectrometry.

METHODS

Surface-trap in-situ detectors, so-called retro-detectors, are based on the track-etch technique. The alpha background activity of a surface may vary and the corresponding background tracks must be distinguished from the $^{210}\text{Po}$ signal (5.3 MeV). Several different principles for this are conceivable,
for example:

- Tandem track-etch detectors of different energy sensitivities
- Discrimination of alpha background emission by foils
- Energy discrimination by track characterisation.

The advantage of all three techniques is that the back of the sample, exhibiting pure background activity, need not be assessed. The tandem method has been developed by SSI, and is described in more detail elsewhere (Falk et al. 1996). In short, this double detector approach exploits the fact that the CR-39 plastic is sensitive to most alpha energies including that from 210Po, while the cellulose nitrate film (LR-115) “sees” only the energy window 1.2-4.8 MeV. By autographical exposure of CR-39 and LR-115 films side by side on glass sheets with known surface activities of 210Po and on background glass samples, calibration coefficients for converting track densities to a 210Po surface activity have been established.

The glass sheets used as calibration pads were chosen to cover a suitable range of implanted 210Po activity. The approximate 210Po level is first obtained by a PIC measurement. The relative surface distribution of the implanted 210Po is then mapped by covering the whole surface with retro-detectors. If selected, the glass sheet is analysed in more detail and then used repeatedly for calibration and constancy checks of retro-detectors, as well as the pulse-ionisation chamber system.

Several methods have been applied for the activity analysis of the glass sheets used as calibration pads:

- alpha spectrometry of the glass surface by a pulse ionisation chamber
- alpha spectrometry of the glass surface by surface barrier detectors
- gradual removal of the glass surface activity by etching
- activity analysis of the whole glass matrix by radiochemistry methods.

The flow-through pulse-ionisation chamber (PIC) is shown schematically in Figure 1. The PIC accommodates semi-infinite samples on top and is used for the nondestructive determination of low- 210Po-activity glass sheets (Johansson et al., 1992). The detector can be collimated to opening diameters between 3 and 26 cm. For diameters less than 20 cm, the alpha peak counting efficiency of the PIC detector is 40% or higher. This efficiency has been determined by scanning the opening diameter of the detector with calibrated point sources. Complementary alpha spectrometry on a few calibration glass sheets was performed with surface-barrier detectors (Ortec DIAD). The effective glass area seen by these detectors is 7.1 cm². The DIADs were individually calibrated at SSI by means of a combined delayed gamma and alpha spectrometry on air-filter samples of radon progeny (Falk et al., 1994). The alpha peak counting efficiency of the DIADs is about 22% over a solid angle of 4π.

In order to reveal the depth distribution of the implanted 210Po activity, the surface of the glass sheets is etched away in steps of about 10 nm. This work is only in its infancy and the results are too preliminary to be reported here. The glass matrix analysis starts by dissolving the glass in hydrofluoric acid. The glass solution is then analysed with regard to all relevant alpha emitters of the thorium and uranium decay series, using standard radiochemical techniques followed by alpha counting, either using ion-implanted detectors or ZnS flasks. Once the alpha activity per unit volume of the glass has been determined, the expected alpha fluence rate at the glass surface is calculated as a function of alpha energy using the continuous slowing down approximation.
RESULTS AND DISCUSSION

Due to the large area seen by the PIC detector it is possible to extend the sensitivity to below 1 Bq/m² of implanted $^{210}$Po (see Figure 2). The sample surface outside a radius of 10 cm contributes very little to the alpha peak count rate and consequently, the maximum sample area analysed in practice is 314 cm².

The peak alpha counting efficiency of a PIC detector must be determined by a point-source scanning technique, as no large-area reference alpha sources are commercially available. The efficiency falls off towards the periphery of the detector opening, and from the shape of this efficiency curve, the peak counting efficiency for a large source covering the whole opening can be calculated (Figure 3). The DIAD and PIC detectors were compared by analysing the same region of two glass sheets with an even surface distribution of implanted $^{210}$Po of approximately 200 Bq/m². The estimates differed by less 2% for both sheets, a discrepancy within the estimated error of measurement.

A normalised alpha spectrum of a glass sheet used in our calibration exercises (see Figure 5 below) is shown in Figure 4. The $^{210}$Po peak is very distinct, but the number of pulses outside the peak dominates in comparison, as the peak is narrow and the $^{210}$Po activity is low. Pulses below the full energy peak in Figure 3 originate from:

- alpha emission from $^{210}$Po ionising outside the full-energy region of the PIC
- alpha background activity of the glass matrix
- alpha background activity residing in the interior of the PIC
- electronic noise

When comparing spectrometric results from PIC measurements and track densities obtained by retro-detectors on the same glass sample the distinction between these sources is necessary. Obviously, the retro-detector responds only to the first two categories, but the artefact of the PIC detector, i.e. scattering $^{210}$Po alpha particles below the full-energy peak, has no direct counterpart in the retro-detector response. The fact that the PIC detector combines $^{210}$Po alpha and glass background pulses in the region below the full-energy peak is a problem that we address in two main ways:

- measurements on the front and back of the same glass sheets, with no $^{210}$Po implanted on the back
- experimental determinations of all alpha emitters in the glass matrix by radiochemical methods.

The glass matrix activity analysis is far from complete, but it appears from our preliminary results that low-background vitreous materials contain typically ten millibecquerels per g of alpha emitters of the thorium and uranium series. Such an activity level corresponds roughly to a PIC detector count rate of 100 cph above 2 MeV, assuming a sample diameter of 10 cm.

Calibration data obtained for the SSI retro-detector are shown in Figure 5. Only the critical low-activity range, approximately 1-8 Bq/m² of $^{210}$Po, is shown. Despite the minimal $^{210}$Po activity involved, and the variation in alpha background of these sheets, the dispersion of the data is low. The relative standard deviation of the calibration coefficient determination is about 10%.
CONCLUSIONS

The ability of pulse-ionisation chambers (PIC) to nondestructively reveal the low activity of $^{210}$Po implanted into large-area objects (not necessarily glass sheets as in this paper) is a great advantage when calibrating retro-detectors based on track-etch techniques. It is clear that the alpha emission rate, from both background and $^{210}$Po, of any selected reference sheet should be characterized in detail, in order to achieve a successful calibration. By repeatedly comparing the $^{210}$Po response of SSI retro-detectors and a reference PIC spectrometer, we have verified that the track-etch retro-device is able to measure implanted $^{210}$Po in the low-activity range of 1-8 Bq/m², with a relative standard deviation of ± 10%, under laboratory conditions.

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REFERENCES


Figure 1: A schematic diagram of the open-flow pulse-ionisation chamber (PIC). The sample on top constitutes the cathode. The Frisch grid and anode are made of stainless steel and the chamber walls of plastic, electrically shielded on the outside with aluminium.
Figure 2: Measurement of $^{210}$Po alpha activity implanted into a household glass sheet. The net peak alpha count rate (counts per hour, cph) is given as a function of sample collimation diameter. The count rate is normalised to an activity of 1 Bq/m$^2$. 

![Graph showing the relationship between sample diameter and P/cph count rate.](image)
Figure 3: Peak efficiency of the PIC detector obtained by scanning the circular opening (diameter = 10 cm) with a $^{230}$Th point source. The resulting peak counting efficiency for an extended, homogeneous alpha source is 41.8 \%.
Figure 4: Alpha spectrum, normalised to a counting time of 1 hour, obtained with the PIC collimated down to a diameter of 10 cm. The $^{210}\text{Po}$ implanted activity of the glass sample is 3 Bq/m$^2$. The polonium peak, with a full width at half maximum (FWHM) of 42 keV, is found at 5.3 MeV, and the measuring time is 15 hours and 52 minutes. The peak in the lower part of the spectrum is due to electronic noise.
Figure 5: The SSI retro detector response to 7 low-activity glass sheets. The activity on five of the glass samples was measured twice. The activity of the implanted $^{210}\text{Po}$ nuclei varies between 1 and 8 Bq/m².

| Sensitivity $(\text{tracks/cm}^2\text{h}) / \text{Bq (surface activity)}$ |
|---|---|---|---|---|---|
| 0.0  | 2.5  | 5.0  | 7.5  | 10.0 | |
| 0.00  | 0.02  | 0.04  | 0.06  | 0.08  | 0.10  | 0.12  |

Mean $0.081 \pm 0.008$ SD