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PLATE-OUT AND IMPLANTATION OF ^{222}Rn DECAY PRODUCTS IN DWELLINGS

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Airborne radon (^{222}Rn) decay products deposit onto all open surfaces in a dwelling. A fraction of the deposited short-lived progenies will be immobilized due to alpha recoil decay implantation. The implanted activity of long-lived radon progenies has two different areas of application, either as a retrospective radon exposure monitor or as a long-term tracer for submicron particle deposition processes. To reveal the surface homogeneity of implanted activity, glass sheets from a dwelling were scanned by collimated pulse ionizing chambers. The scanning of individual glass sheets revealed that the implanted activity is homogeneous within approximately ± 20% over the surface. The surface activity frequently decreases close to the bordering frame, indicating a disruption of the airflow boundary layer. In a pilot study involving 23 detached houses of similar type, the implanted activity of the glass objects correlated well with the estimated radon exposure (r^2=0.55). In one home, the ingrowth of implanted ^{210}Pb in four initially unexposed picture frames was followed for 3 y. In the two deliberately chosen recessed places, the plate-out rate was approximately 40% lower compared to the two openly exposed objects. Larger variations in plate-out rates should be expected near radiators and other airflow generators.

INTRODUCTION

When, in 1987, in a dwelling experiment, efforts were made to decontaminate glass sheets from deposits of short-lived radon (^{222}Rn) decay products (SRnD), they were only partially successful. ^{218}Po was easy to remove completely, but a significant proportion of ^{214}Po 'stuck' to the surface and could not be removed, even with elaborate cleaning methods. This phenomenon of the 'uncleanable' glass had also puzzled the radioactivity pioneers, e.g., Crookes (1903), but had already been correctly explained in 1909 as alpha recoil implantation of the daughter nucleus (Hahn and Meitner 1909). The objective of this paper is to describe the experience using superficially implanted activity for the purpose of radon monitoring in retrospect and as fingerprints of particle deposition processes in the indoor environment.

Evidently, only the implanted long-lived radon daughters (LRnD), starting from ^{210}Pb ($T_{1/2}$ = 22 y), are of interest for retrospective radon exposure estimates. Due to the low activities of LRnD involved (typically of the order of a few becquerels per square metre), it was found necessary to calculate ^{210}Pb from alpha spectrometry measurements of its granddaughter, ^{210}Po. An open-flow pulse ionization chamber accommodating semi-infinite glass samples undestructively was developed for the purpose (Johansson et al. 1992). The implantation depth of about 0.1 µm in glass for the recoiling nucleus means that alpha self-absorption is small. The latest ionization chamber has an energy resolution (FWHM) of the alpha peak from ^{210}Po better than 35 keV for a 12-h measurement and chamber openings up to 20 cm in diameter.

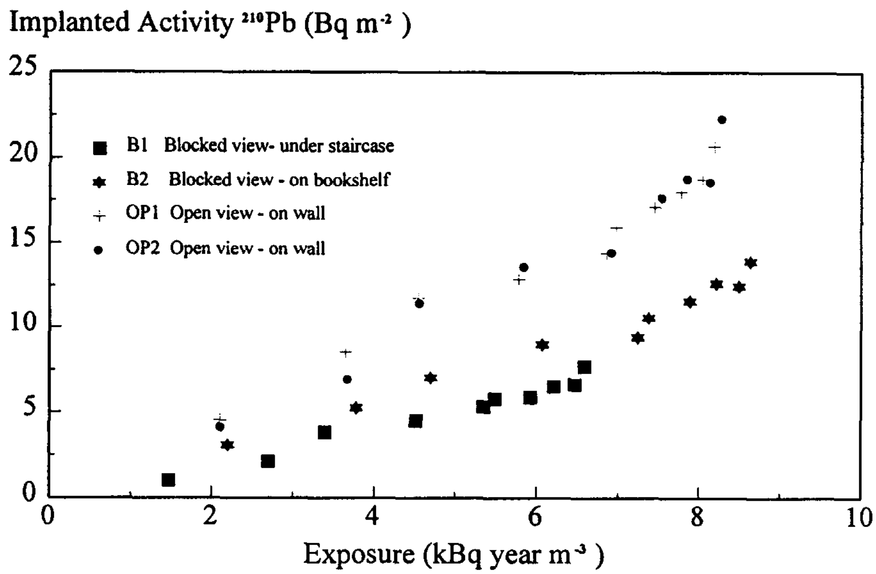

Fig. 1. The growth of implanted ^{210}Pb in four different framed glass sheets exposed in a radon dwelling for 3.5 y. Each point corresponds to a measurement of implanted ^{210}Po in the laboratory and an evaluation and exchange of the radon track-etch detectors exposed at the position of each glass sheet.

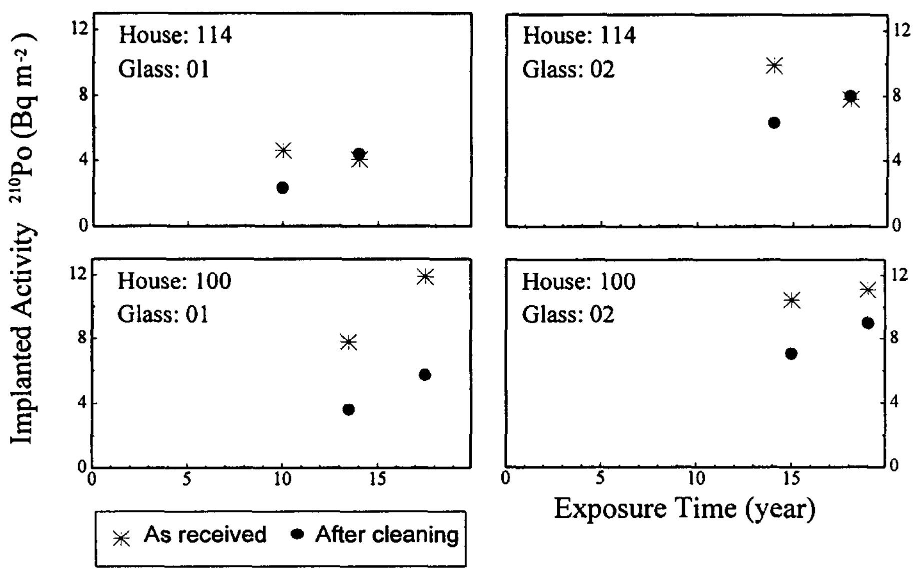

Fig. 2. Four individual glass samples from two different dwellings analysed at two occasions four years apart.

The small implantation depth favours high-resolution alpha spectroscopy but raises questions of durability and interference from dirt on the surface. It is only in hard materials such as metals and glass that long-term migration of the implanted activity can be expected to be low enough to be substantially 'slower' than the half-life of ^{210}Pb. All experience from glass measurements supports the hypothesis that the removal of implanted LRnD is only caused by radioactive decay. Normal glass-cleaning procedures will not influence or remove the implanted activity. The results illustrated in Figs. 1 and 2 support this conclusion. The four glass samples of Fig. 1 were followed for 3½ y and were thoroughly washed in the laboratory 3-4 times a year, prior to each measurement. Two of the samples (B1 and B2) were deliberately placed in recessed locations.

The results illustrated in Fig. 2 from two different detached houses suggest that some objects were cleaned by the owners just prior to the analysis. There is about 4 y between the two measurements shown in Fig. 2, and an important observation is that the growth of the implanted activity seems to follow the same trend as during the first 10-15 y, despite differences in cleaning habits of the households. On some objects, the removable LRnD activity exceeds 50%. This does not necessarily prove that a significant implantation of alpha recoils takes place in the surface contaminant itself, rather than in the solid glass. One reason for this is that the dust landing on the glass surface may be of different age and therefore of different LRnD concentration. This idea is speculative, but as some of the samples are positioned beneath open staircases, the resuspension and fallout of old dust can not be excluded.

The experience described here regarding the durability of the implanted activity is limited to glass samples exposed at room temperature under dry indoor conditions. The conclusions presented on implant integrity can not be applied to high-humidity or water-condensing conditions, under which surface corrosion of glass, especially at elevated temperatures, may be significant.

Even if the interference from dust and grease is of little significance for the samples in Fig. 2, the possibility that in certain indoor environments surface contaminants may have a deleterious effect cannot be excluded. Systematic studies on how household contaminants on surfaces interfere with the implantation process are lacking. The results from a pilot study of household glass sheets exposed in a walk-in radon chamber indicate that a significant fraction of implantation can take place in the surface dust (Johansson et al. 1994). The removable fraction of ^{210}Po after exposure was 32 and 49% for clean and dusty samples, respectively, but the spread in the individual results of the 20 samples was large. The results from this pilot study support the hypothesis that indoor surface contaminants can significantly reduce the implantation rate of LRnD into the glass matrix, though the reduction is much less than expected from pure range/mass-load calculations, due to the uneven surface distribution of the dust.

The implanted ^{210}Pb activity originates from the SRnD deposited on the surface and this 'source strength' can vary substantially in the indoor environment due to aerosol, airflow, and geometry conditions. Upon consulting the literature on indoor deposition velocities of SRnD, differences of the order of two magnitudes are found. Fortunately, the extreme experimental conditions behind some of these results do not exist in dwellings, and short-term and seasonal fluctuations of plate-out rates are smoothed out due to long integration times. The results shown in Fig. 1 exemplify the variation in long-term plate-out rates in a case where radon exposure and cleaning habits are fully controlled. In the two deliberately chosen recessed locations (B1 and B2), the implanted ^{210}Pb activity is only 40% less than on the two openly exposed objects. Larger variations in SRnD plate-out rates could, however, be expected near radiators and other airflow generators.

Local plate-out variations are a source of error which must be dealt with, and as reliable correction factors for geometry, airflow, and aerosol conditions are not available, the best recipe for improving the correlation between radon exposure and implanted LRnD activity is to analyse several glass samples from each dwelling. The possible errors introduced by surface contaminants also speak in favour of a multi-sample approach. Fortunately, large-scale multi-sample investigations are now feasible thanks to the development of cheap track-etch devices suitable for *in situ* measurements of implanted ^{210}Po (Falk et al. 1996; Fitzgerald 1996).

In one-sample-per-house measurements, the correlation between radon exposure and implanted activity can be improved using samples that have been exposed in a similar fashion and excluding certain categories of samples, e.g., windows and samples greasy from exposure in kitchen areas. In a one-sample-per-house study of 23 detached houses of similar age and construction, the implanted ^{210}Pb activity of framed glass objects correlated well with the estimated radon exposure (r^2=0.55), considering the questionable validity of the radon exposure estimate, based on contemporary radon measurements by radon track-etch detectors exposed during a two-month period. (The radon exposure values were estimated by multiplying the track-etch radon concentration value by age of exposure as given by the owner).

The implanted LRnD activity technique can be applied in areas other than the indoor retrospective radon monitoring discussed above. Figure 3 shows an example of how the implanted activity can be used to reveal the pattern of small-particle deposition processes over long periods of time. Such investigations can be of interest to researchers in the field of indoor aerosols and indoor pollutants. The more detailed surface activity distribution was obtained by scanning the surface of the glass sheet with a pulse ionization chamber collimated to a slit width of 5.3 x 138 mm^2 (Fig. 3a) and a diameter of 30 mm (Figs. 3b-d), respectively. The slit collimator (Fig. 3a) improves the spatial resolution in the vertical direction and a decreased plate-out rate close to the horizontal part of the wooden frame is revealed, an effect presumably due to the disrupted boundary layer.

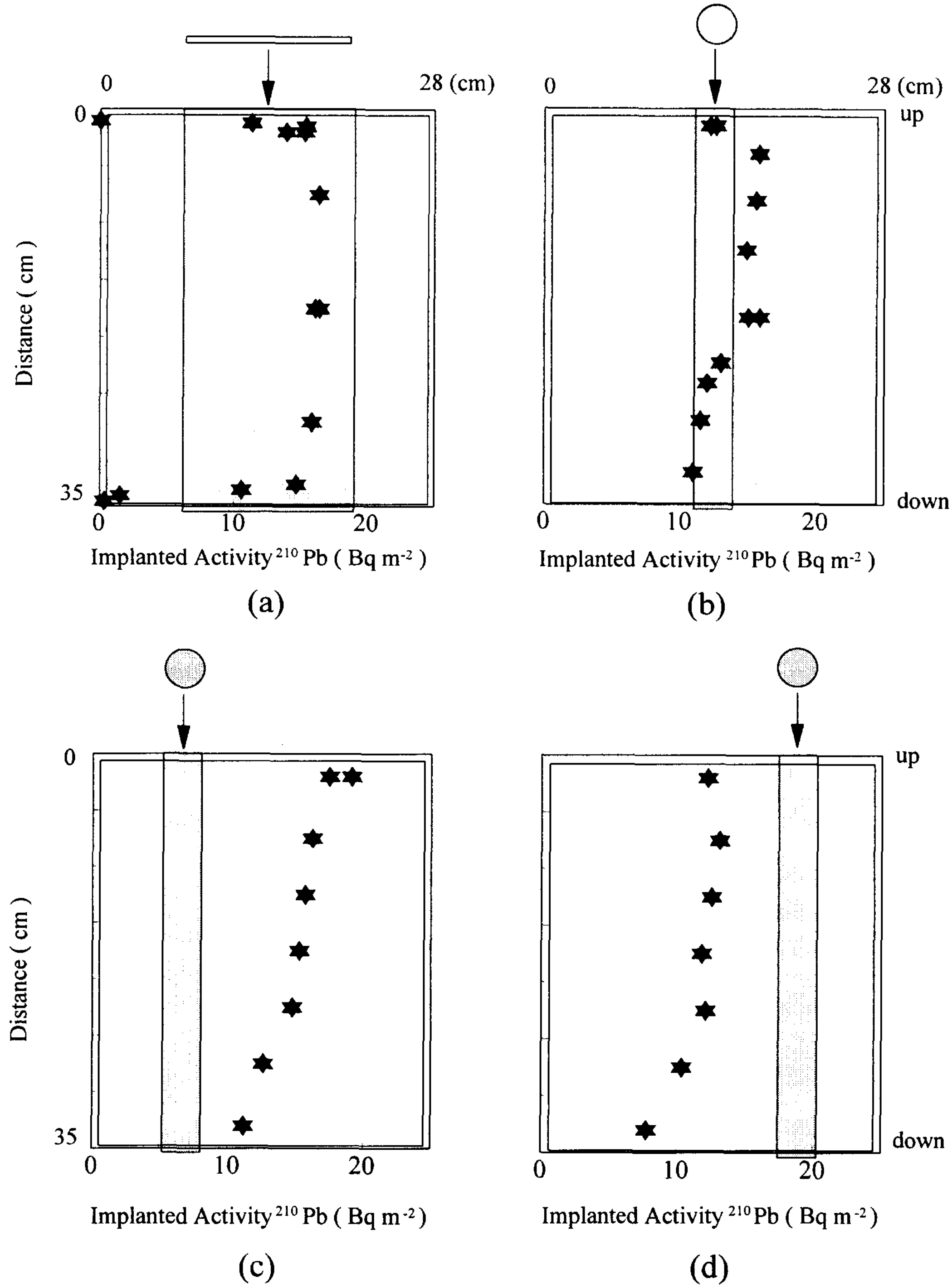

Fig. 3. Results from an activity scan of sample OP2 (a) and B2 (b-d) of Fig. 1. The slit collimator (a) is of the same area as the circular one (b-d) and the scan position is indicated by the shadowed area.

As can be seen from Figs. 3b-d, the implanted activity is distributed fairly homogeneously over the framed glass, despite the recessed position above a bookshelf with the frame leaning against the wall and close to the ceiling. There is a tendency towards less activity implanted in the lower half of the glass sheet, which may be a shadowing effect of a vase in front of the frame.

It is obvious from the examples above that several factors can interfere with the application of implanted activity as a retrospective radon monitor. Through a careful choice of samples and a multi-sample approach, the correlation between radon exposure and implanted activity is expected to be good within a factor of 2. With a limited budget, there are no alternative methods which can quantify radon exposure in retrospect and that can be applied on a large-scale basis. The advantages of being generally applicable, highly specific to airborne activity, and the availability of non-destructive *in situ* detectors, cannot be matched by any other technique.

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