DOE/ER/60911--4

DE93 010463

**^{210}Po as a Long-Term Integrating
Radon Indicator in the
Indoor Environment**

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Final Report:
SSI Project No. P505.88
DOE Project No. DE-FG02-89ER60911

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BACKGROUND

Exposure to radon (Rn-222) decay products in the indoor environment is suspected of being a significant lung cancer agent in many countries. But quantification of the contemporary lung cancer risk (i.e. probability) on an individual basis is not an easy task. Only past exposures are relevant and assessing individual exposures in retrospect is associated with large uncertainties, if possible at all. In the lung cancer time perspective all contemporary radon and radon daughter measurements, even if extended over, say, a one-year period, should only be considered as more or less relevant grab samples.

One way to extend the validity of contemporary measurements to past decades is to measure long-lived decay products of radon, the long-lived radon daughters (LRnD). After our laboratory had exemplified the correlation between implanted Po-210 and the estimated radon exposures in six different dwellings⁽¹⁾, the US Department of Energy (DOE) and the Swedish Radiation Protection Institute (SSI) granted funds for a one-year study, "²¹⁰Po as a Long-Term Integrating Radon Indicator in the Indoor Environment" (DOE grant No. DE-FG02-89ER60911, SSI Grant No. P505.88). In this report the work performed under these two contracts is reported.

OBJECTIVES AND APPROACH

The objectives of the project were to determine the correlation between airborne radon/radon daughter activities and the implanted activity of hard surfaces such as glass, to identify factors in dwellings affecting this correlation and to take a closer look at the surface plate-out/implantation mechanism.

The problems posed have been investigated by means of small-scale pilot studies. The limited budget has not permitted any large-scale involvement of personnel or dwellings. In order to facilitate *in situ* measurements of Po-210 and short-lived radon daughters (SRnD) part of the SSI grant was used to finance a large (40 cm²) surface barrier detector (SBD). In order to be able to analyse Po-210 on glass surfaces without dismantling or cutting the glass into pieces, it was necessary to develop a special type of pulse ionization chamber (PIC).

RETROSPECTIVE METHODS

Before describing the studies in more detail it is worthwhile to briefly consider the retrospective methods available for radon exposure studies.

Measurement of radon or short-lived progenies in homes

One approach is to estimate the exposure to a person by measuring radon or radon daughter concentration levels in one or several dwellings in which the person has lived and multiply the obtained values by the fractional times spent indoors. This is the best way to estimate exposure in retrospect, providing that all, or at least most previous homes can be traced, the radon/radon daughter concentration measured and that the length of time spent indoors at each place is available. Obviously, this multi-home approach is very laborious and costly. Monte Carlo simulations of typical Swedish conditions⁽²⁾ reveal a high correlation ($r^2 = 0.65$) between estimated and true exposure only if all the homes occupied by a person are taken into account. The radon value of today in the last home of a typical Swede is, on the other hand, both biased and poorly correlated to the true exposure (-0.40 , $r^2 = 0.11$).

Measurement of long-lived radon daughters

The activity of the long-lived decay products that radon leaves behind in a house reflects past radon exposures due to the long half-life of Pb-210 ($T_{1/2} = 22$ y). It is only when this buildup of activity can proceed undisturbed over many years, that we have a system which is potentially useful for retrospective radon/radon daughter exposure studies. To the radon epidemiologist the measurement of LRnD is very attractive since the time scale for the buildup of lung cancer risk and Pb-210 activity are assumed to be approximately similar. LRnD in objects that move with person constitute the ideal retrospective radon monitor from an individual point of view, but personal objects suitable for retrospective LRnD studies are rarely available. The normal case, as with SRnD investigations, is that the measured level concentration characterizes the house and not the individual.

Three different classes of traps accumulating long-lived radon daughters (LRnD) in houses are of interest:

1. humans
2. volumes
3. surfaces.

Humans

Utilizing the LRnD retained by the human body for retrospective radon studies is attractive because the measured value is directly coupled to the individual and not to a house. Unfortunately, the residence times of most organs for LRnD are short and other intake routes besides inhalation of short-lived daughters are significant. The residence time of Pb-210 in the skeleton may be long enough, but on the other hand, bone samples can not be taken from living people.

For highly exposed miners ($> 30 \text{ kBq y m}^{-3}$ radon gas) a clear correlation between Pb-210 in bone and radon exposure can be seen⁽³⁾⁽⁴⁾ but the contribution from the inhalation of uranium dust is not known. A comprehensive literature survey of Pb-210 levels in humans as a radon exposure indicator has been published by Enflo⁽⁵⁾.

Volumes

A volume trap must give free entrance to radon gas while retaining the LRnD with high efficiency. Spongy objects, unpainted wood or dust in normally inaccessible places may fulfil the requirements for useful traps, but if only small volumes are available, sensitivity may be a problem. Interior volumes of mattresses, furniture etc. consisting of foamed plastics are potential candidates as useful traps, but to our knowledge no papers describing these as indicators have been published so far. If expensive objects in the dwelling are the targets for LRnD monitoring, the destructive nature makes radon retrospective monitoring involving volumes costly and therefore limited to small-scale studies.

The Pb-210 content of dust sampled from 12 dwellings in Sweden has been investigated by Forberg⁽⁶⁾. A correlation between Pb-210 activity and the measured radon concentration was found, but the data were widely scattered. The usefulness of dust as volume traps for LRnD is also limited by the fact that the age of the dust is not well defined.

Surfaces

A substantial fraction of the airborne short-lived decay products of radon is removed from the air by deposition processes. All indoor objects and surfaces are thus contaminated by SRnD. The deposition of SRnD is a complicated process influenced by particle size, surface orientation, air movements etc. In order to avoid local effects of air streaming, turbulence, and shadowing, all phenomena difficult to quantify, only openly exposed surfaces are recommended as LRnD traps. Open surfaces are also easy accessible for cleaning and other superficially destructive measures, implying that the LRnD residing loosely on objects in a room are not very useful for retrospective radon studies. Fortunately, a fraction of the deposited atoms implant their daughter atoms by means of alpha recoils. The implanted activity resists all normal cleaning procedures and forms an ideally thin alpha/beta source. The range of the alpha recoiling nuclei in glass surfaces is about $0.15 \mu\text{m}$ ⁽⁷⁾. The alpha recoil is a key process concerning surfaces as LRnD traps. The alpha recoil phenomenon was explained for the first time in 1909 by Meitner and Hahn⁽⁸⁾, and reintroduced into radon research in 1988 by Samuelsson⁽¹⁾.

One attractive feature of surfaces as LRnD traps is that objects suitable for measurement can

be found in all homes and that the measurement can be performed non-destructively. The most serious drawback is that the intermediate process of SRnD plate-out varies with many non-dose parameters. Only objects exposed under similar conditions should be compared. A good correlation between radon exposure and LRnD surface concentration can easily be obscured by large variations in plate-out.

Radiation damage

Radiation damage is also a consequence of radon/radon daughter irradiation of indoor objects. In certain materials the microscopic damage done is stable enough to allow retrospective evaluation. Unfortunately, the principle is not generally applicable as only special materials are useful. Fleicher⁽⁹⁾ has suggested track-etch analysis of CR-39 plastic in sun-glasses as a retrospective radon method.

DETECTOR DEVELOPMENT AND APPLICATIONS

Initial Considerations

Analysing the LRnD surface activity on authentic glass sheets imposes problems due to the low activities normally involved. A radon exposure of 1000 Bq y m^{-3} (27 pCi y l^{-1}) implants roughly 1 Bq/m^2 (2.5 pCi/ft^2) of Pb-210. Pb-210 is an almost pure beta emitter and difficult to analyse in the low-activity region. The sensitivity and specificity are greatly enhanced if the alpha-decaying grand-daughter of Pb-210, Po-210 is analysed instead. If the radon exposure history of an object is known it is easy to convert from Po-activity to Pb-210 activity.

At the beginning of the project the choice of glass material taken from homes was restricted to fairly small and inexpensive objects. The reason for this was that the glass sheets had to be cut to a diameter less than 180 mm to fit into the closed pulse ionization chamber available at that time. This destructive feature prohibited, or at least made it expensive, to analyse window glass, large mirrors, glass from cupboard doors etc. taken from dwellings. One of the primary tasks of the project was therefore to develop non-destructive methods for alpha spectrometry of Po-210.

Non-destructive alpha spectrometry of Po-210

Surface barrier or ion-implanted semiconductor detectors are a standard choice for low-level alpha-particle spectrometry, but the small size of standard detectors, a few 100 cm^2 , is a disadvantage when analysing large samples of low areal activity. A parallel plate pulse

ionization chamber (PIC) is more adapted to large-area alpha spectrometry and this type of detector has been used throughout the project as a reference detector.

The development of pulse ionization chambers

Only one pulse ionization chamber (Fig. 1) was available initially. The chamber was built in 1987 with support from an Australian laboratory⁽¹⁰⁾ and is similar to the detector presented by Hötzel and Winkler⁽¹¹⁾. The alpha energy resolution (FWHM) and counting efficiency (2π) of implanted Po-210 samples of maximum diameter (180 mm) are 70 keV and 0.65, respectively. Data and settings of the closed chamber are given in Appendix A. The weight of the chamber makes it impractical to move and during the project the chamber has essentially been used as a stationary detector in the laboratory.

Fig 1. A schematic drawing of the closed parallel plate pulse ionization chamber.

A closed type of PIC limits the sample diameter and this is definitely a disadvantage when dealing with authentic and perhaps valuable glass samples from dwellings. One of the goals of the project was therefore to develop a new type of ionization chamber meeting the following requirements:

- i) alpha energy resolution and counting efficiency for large-area samples comparable to closed PICs
- ii) capability for non-destructive measurements of plane semi-infinite samples
- iii) a non-stationary detector that can be used for field and *in situ* studies.

Fig 2. A schematic drawing of the open-flow pulseionization chamber.

All three goals were accomplished by an open-flow chamber made of polypropylene (Figure 2). This type of PIC is used in an "upside-down" geometry and sample changing is thus facilitated. The glass object to be measured constitutes the cathode of the system. Detailed data and settings of the first constructed open-flow counter are given in Appendix B. Later, an open-flow PIC with a larger diameter opening and improved energy resolution was developed. This later generation of chambers has been described in some detail by Johansson et al.⁽¹²⁾. Unless stated otherwise, the data given in the following text refers to the improved version of the pulse ionization chamber.

Fig 3. Alpha spectrum obtained with the improved open-flow pulseionization chamber.

In the first construction, five thin wires of stainless steel just beneath the glass surface helped define the cathode area, but it was soon found that the chamber behaviour was not impaired

if these wires were removed. The energy resolution of implanted large-area Po-210 sources was about 60 keV (FWHM) in the first plastic chamber, but in later chamber constructions this value was reduced to less than 40 keV (Figure 3). Both the counting efficiency and the energy resolution are insensitive to changes in counting gas flow rate (cf. Figures 4 and 5). The open chambers are of a rigid construction and accommodate heavy samples, for instance large mirrors, without extra support. The maximum opening of the chamber is 22 cm. The alpha counting efficiency drops to 50% (2π geometry) at 10 cm from the centre of the

Fig. 4. The counting rate of a ^{233}U point source as a function of counting gas flow rate.

Fig. 5. The alpha energy resolution of the first open-flow chamber as a function of gas flow rate.

sample. Compared with a 450 mm² surface barrier detector the net count-rate from identical surface activity densities for the open-flow chamber is a hundredfold higher.

The counting gas fed to the chamber is of high purity and no drying agent inside the chamber is necessary. Long-term measurements overnight or longer are possible without any significant deterioration in energy resolution. A gas flow regulator keeps the gas flow constant irrespective of the pressure in the chamber volume.

Due to the plastic body the open-flow chambers are lightweight (5 kg) and mobile. The chambers built so far have mainly been used in the laboratory for measurements of glass samples brought in from dwellings. A chamber has been used in the field on a few occasions. During field exercises the necessary equipment was installed in a small mobile home that was parked outside the dwelling to be investigated. Both SRnD and LRnD could then be investigated on windows, glass cupboard doors, and mirrors taken from the house, due to the short decay times before measurement.

Fig 6 The open-flow pulseionization chamber measuring Po on a vertically oriented window in the laboratory.

The weight and need for a continuous gas supply make the open pulse ionization chambers awkward for *in situ* measurements of Po-210 but, as illustrated in Figure 6, the chamber can, if necessary, be attached to vertical objects. The weight of new constructions can easily be reduced to say 1-2 kilograms, but even then, equipment size and costs prohibit any large-scale *in situ* use of pulse ionization chambers.

The utilization of the closed ionization chamber has now decreased in favour of the open-flow type. Most of the Po-210 measurements presented in this report have been performed with the open-flow type of detector.

A large-area surface barrier detector

Besides the PIC detectors, a large-area (4000 mm²) surface barrier detector (SBD) has also been used for implanted Po-210 spectrometry (Figure 7). (The SSI part of the funding financed this rather expensive detector). The large SBD was used in the early stage of the project, mainly for pilot *in situ* investigations, and has as such been a very valuable complement to the PIC detectors.

Fig 7 A schematic drawing of the large surface barrier detector (SBD) mounted in the aluminium vacuum chamber. The detector area is 40 cm²

Compared with PIC detectors, the SBD has a lower sensitivity to areal sources of Po-210 (due to the larger source-to-detector distance and a smaller detector surface) but is more reliable and wins in speed and versatility over the PIC. The SBD is however sensitive to contamination, especially when used in vacuum. To prevent the buildup of long-lived alpha contaminants the detector was shielded by a 1.5 μ m thick Mylar foil during most measurements. The foil increases the alpha energy resolution from typically less than 90 keV (FWHM) to about 300 keV for a small Am-241 source.

Fig 8a The leakage current of the large surface barrier detector as a function of detector temperature.

Fig 8b The alpha peak shift of the SBD as a function of bias current.

The production of electron-hole pairs in a semiconductor is temperature dependent and this temperature dependence gives a peak drift which causes the energy resolution to deteriorate if the temperature varies during a measurement. Figure 8 shows how the bias current changes with temperature and the corresponding peak shift. The temperature dependence in combination with a small (the detector is of the ruggedized type) but significant sensitivity to light make the SBD less suitable for *in situ* measurements on windows.

A special aluminium housing for the SBD detector was built to facilitate measurements on glass both in the laboratory and *in situ* (cf. Figure 7). The specifications and electronic equipment of the SBD are summarized in Appendix C. It is apparent that the large area, to some extent, spoils the good alpha energy resolution typical for smaller SBDs.

RESULTS AND DISCUSSION

Plate-out and implantation

The deposition of SRnD onto surfaces is a critical process in the transformation chain from airborne to implanted activity. In general the plate-out rate varies with aerosol conditions, surface temperature, surface position and orientation, air streaming etc. Radon room experiments and theoretical calculations indicate that the impact on SRnD plateout from non-dose correlated phenomena is significant. A completely free choice of exposure conditions and type of sample will obscure the relationship between potential lung dose and implanted activity.

The deposition of SRnDs is a "source" for the implanted activity and it is essential to study how this source strength varies locally on a small (cm) and large (m) scale and with exposure conditions. Measurements of the implanted LRnD at different locations in a house will reflect the long-term spacial variations in plate-out rate providing the airborne radon daughter concentrations have remained constant.

Another matter of importance is the alpha recoil implantation process. The recoiling nucleus has a range in glass about $0.05 \mu\text{m}^{(7)}$. Thus, it can be suspected that a significant part of the total number of recoils on dirty surfaces will not be able to penetrate into the glass matrix. LRnD activity residing in the loose and easily removed contaminants of a surface is of no value for long-term retrospective monitoring. The fate of the alpha recoils during dust load conditions met in the home environment must be investigated before methodologies for retrospective studies based on implanted LRnD can be formulated.

Recoil implantation

Subsequent to an alpha decay the recoil energy of the daughter nucleus is sufficient for the nucleus to penetrate into the underlying material or be ejected from an aerosol particle or a macroscopic surface (cf. Fig. 9). The range of the recoils in air is very short and most of the nuclei recoiling from a surface will presumably be recaptured in the indoor dwelling environment. The part of the deposited SRnD which is implanted by means of alpha recoils

Fig 9 Initial velocities and energies for the alpha decay of Po-218.

into the glass matrix will be non-removable unless the outer surface of the glass is etched. Before the recoil effect was explained by Hahn and Meitner⁽⁸⁾ in 1909, the non-decontaminable glassware syndrome was very confusing for scientists working with radium, "emanation" (radon and thoron), and the "active deposit" (daughter products). Crookes⁽¹³⁾ formulation from 1903 is a good example:..." the persistence of radio-activity on glass vessels which have contained radium is remarkable. Filters, beakers, dishes used in the laboratory for operations with radium, after having been washed in the usual way, remain radio-active..."

In earlier days before electrolytic deposition techniques were established the alpha recoil effect was frequently used for the production of thin alpha sources.⁽¹⁴⁾ The implantation effect was in most cases enhanced by electric field acceleration and by the use of evacuated implantation vessels. Since the introduction of the surface barrier detector and alpha spectrometry of samples in evacuated chambers the alpha recoils have been problematic to the scientist analysing low-activity specimens from the uranium series. The slowly increasing alpha background due to implantation in the detector surface cannot be cured by cleaning even if the detector is of the cleanable type.

Lively and Ney⁽¹⁵⁾ considered only diffusion as a transport mechanism for the penetration of SRnD into the underlying material, and were apparently not aware of the alpha recoil phenomenon. They suggested glass as a suitable substrate material because depth penetration due to diffusion is negligible and consequently the alpha self-absorption of the deposited activity will remain insignificant for decades. So what Lively and Ney actually suggested was a retrospective radon system that relies on surfaces "never touched". Dwellings with openly exposed glass surfaces that have been left undisturbed for several years must be very rare, and it is in fact the hiding away effect, caused by alpha recoil implantation, that makes the glass-polonium system applicable in almost any home. The excellent idea of using LRnD alpha activity on glass surfaces to assess average long-term radon air concentration levels, introduced by Lively and Ney⁽¹⁵⁾, was apparently of much greater potential than the authors were aware of themselves.

The implantation effect by recoils is easy to demonstrate experimentally in dwellings. In this project we set up the large SBD detector in a garage with low radon concentrations in the immediate vicinity of a dwelling with radon levels of about 500 Bq/m³. After exposing new glass sheets to SRnD plate-out for an hour in the house, subsequent cleaning could only remove about half of the surface activity of Po-214 (RaC'). This implies that a very rapid process must be responsible for binding the unremovable part of the RaC' activity to the surface. Recoil implantation is such a rapid process and the fact that half of the activity could

be removed indicates that the dominating activity plating out is Po-218 and that the implantation takes place when this radionuclide emits an outward bound alpha particle.

Plate-out patterns

As vitreous glass in the indoor environment is essentially free from electrostatic charging one would expect the SRnD plateout variations on a cm-scale to be insignificant. (Exceptions may occur close to corners etc. and the edges of the glass sheet). The homogeneity of plate-out on this small scale has been tested by placing large sheets of CR-39 track-etch films on the exterior and interior surfaces of a glassed cupboard door (Figure 10). The radon concentration in the room outside the cupboard and inside the cupboard is the same, but the interior plate-out rate is reduced by roughly 50%. A comparison of the track densities of interior and

Fig 10 Track densities of a bare CR-39 detector exposed openly for a week attached to the outside of a glassed cupboard door. (The evaluation was made by SSI.)

exterior CR-39 sheets, exposed for a week in a dwelling with a level of about 500 Bq/m³ radon gas, indicates that most tracks originate from plate-out of SRnD and not airborne radon or decay products. According to calculations made by McLaughlin⁽¹⁶⁾, the signal from a non-electrostatic bare CR-39 detector from deposited activity is 80% or more during typical room conditions. The results exemplified in Figure 10 give support to the idea that the plate-out rate onto glass surfaces is homogeneous on a small scale.

On a slightly larger scale inhomogeneities in SRnD plate-out are more likely to occur, e.g.

due to variations in air flow caused by corners or nearby objects, but systematic investigations of spatial and temporal plate-out patterns under realistic indoor conditions are lacking. Window panes are subjected to a number of parameters influencing plate-out and these factors may cause excessive variations along a single window surface and between windows. The radon concentration may be low due to ventilators or frequent ventilation, curtains may cause shadowing effects, radiators (enhanced air flow) and low temperature of the glass (thermophoresis) may both enhance the plate-out etc. Bearing in mind these factors it can be questioned if window panes can be used in practice as reliable radon integrators.

An example of plate-out inhomogeneity is given in Figure 11, where four windows from a single family house were analysed in the lab with the open-flow PIC. The implanted Po-210 activity of the upper part of the windows is enhanced by 13-32% compared with the middle section. The variations seen in Figure 11 are presumably due to differences in air streaming, which in turn depends on radiator and window recess geometries. The intra-window variation illustrated in Figure 11 is moderate and does not as such exclude window panes from retrospective radon monitoring.

Fig 11 The implanted Po-210 of four windows from the same dwelling normalized to the centre position.

In-home variations

Extended and systematic investigations of intra-home variations of implanted Po-210 are outside the scope of this project, but in order to demonstrate the feasibility of field measurements with an open-flow pulse ionization chamber the Po-210 variations in a single-family house were studied.

The house is situated in a town 300 kilometres from Lund. The source of radon is the building material (alum-shale-based lightweight concrete) and the same family has lived in the house since it was built in 1967. Energy saving measures were taken in 1979 by the introducing insulation around the windows and extra loft insulation, and it is reasonable to assume that these steps increased the general radon levels in the house. In retrospect the variation in radon levels can not be assessed with any high degree of accuracy and we have therefore, for simplicity, assumed that the contemporary radon results for each room have prevailed for the whole glass exposure period.

The necessary equipment was installed in a camper and parked just outside the house to be investigated. Mirrors, cupboard door glas and most of the windows were brought from the house one at a time to the camper for Po-210 analysis. Fourteen different objects were analysed and the ratio of implanted Po-210 to the estimated radon exposure is presented in Figure 12.

Fig 12 The ratio of implanted Po-210 activity to radon exposure of 14 different glass samples taken from a single family house.

Bearing in mind all the factors that can influence the window plate-out, the spread in the window values in Figure 12 is surprisingly low. The results are also consistent in the sense that all the windows used for ventilation show a lower Po/Rn ratio than the others. Excluding the kitchen door and the windows used for airing, the spread in the window values is only 10%. This unexpectedly low figure can be partly explained by the fact that all the windows and window openings are of the same configuration and that all the windows (except the one in the kitchen door) have identical windowsills and radiators beneath. It is evident that this single house gives no support at all to the view expressed above where the usefulness of windows as radon exposure meters was questioned.

The glass from the picture frames exhibited a lower Po/Rn ratio than the non-vent windows except for the 5-year-old mirror. This general trend of lower Po/Rn ratio is expected due to the absence of radiators beneath the framed glass positions. Why the relatively new mirror

Fig 13a Discharging curves for four lucite sheets during exposure in a dwelling.

in the hall differs having a high Po/Rn ratio is not known, but there can be several explanations. The radon exposure in the hall may be underestimated (radon measurements are all from other rooms), the age of the mirror may be incorrectly remembered by the residents, the mirror can have been exposed prior to purchase, higher than usual plate-out rate due to the nearby radiator, etc. In retrospect it is difficult to pinpoint the right explanation without re-entering the house.

Static electricity

Static electricity can dramatically increase the SRnD plate-out rate as a significant fraction of the airborne SRnD is charged. In order to demonstrate this, lucite sheets (18x24 cm) were charged and exposed in a radon dwelling (500 Bq/m³ radon gas) for approximately 20 minutes. The results in Figure 13a support the established fact that surfaces in a home charged by rubbing will

be neutralized within tens of minutes. After the exposure in the house the sheets were immediately analysed with the large SBD in a radon-free garage nearby.

During exposure the sheets act similarly to a SRnD air sampler with decreasing sampling velocity during sampling and different collection efficiencies for the three daughters. The spectrometry results obtained with the SBD have consequently been evaluated by means of a SRnD filter program. Due to the complex and unknown behaviour of "sampling rates" only relative values can be given for the Po-218 plate-out (Figure 13b). The plate-out rates for Pb-214 and Bi-214 were both insignificant compared with the values for Po-218.

The majority of Po-218 atoms are "born" as positive ions and negatively charged objects are therefore strong sinks for airborne Po-218. The positive sheet attracts less activity as can be seen in Figure 13b, an observation supporting earlier findings by Bigu⁽¹⁷⁾. Plastic or glass objects rubbed with cloths or paper in homes become, in most cases, negatively charged and will apparently be neutralized within a few tens of minutes. This leads to the conclusion that plastic or glass objects that are frequently rubbed, i.e. several times per day, are unsuitable

Fig 13b Po-218 (RaA) activity per unit area as a function of integrated potential during exposure.

as substrates for LRnD implantation due to the variable plate-out rate. Plastic spectacle lenses may be one example of objects that must be excluded for such electrostatic reasons.

It is interesting to note that the household cleaning of glass sheets may affect the level of implanted Po-210 activity in two ways. Firstly, the short-lived activity already deposited is, at least partly, removed, thus taking away short-lived activity that will eventually be implanted by alpha recoils. Secondly, if the surface becomes charged, the SRnD plate-out rate will, for a short while, increase. These two effects of cleaning obviously compensate each other, but it is not known if the result is an increase or decrease in implanted Po-210.

Considering this compensation effect, the short charging times, and that glass is charged positive by the typical cleaning cloth (made from paper, cotton, or linen), a cleaning frequency as high as once a day will have an insignificant impact on the implanted activity of Po-210.

The influence of dirt and dust

Surfaces in a dwelling are more or less contaminated by dust and grease and it is not known what fraction of the recoiling atoms is caught by loose contaminants and what fraction penetrates into the underlying glass sheet. One way to investigate the fate of the recoils is to analyse glass sheets exposed to both contaminants in the lab and SRnD plate-out *in situ*. The basic hypothesis is that 50% of the inward-going recoils are implanted into the glass matrix.

Fig 14 Percent Po-210 activity remaining after wet-cleaning 16 household glass sheets.

If the hypothesis is true and the implanted activity is undisturbed by cleaning, it follows that it will be impossible to remove more than 50% of the long-lived decay products from the surface. This conclusion is not dependent on the probability of surface recapture of the outward-directed recoils. In the project 16 glass sheets covering photos, paintings etc. from 12 different dwellings were analysed in our laboratory for Po-210 before and after careful cleaning. The remaining Po activity is shown in Figure 14 and the conclusion is that none of the glass sheets reject the hypothesis. The test is rather weak though, as the amount of removed contaminants and recoils by household cleaning prior to measurement is unknown.

A more critical test would be to expose authentic household glass samples from non-radon dwellings in a radon chamber under well controlled conditions and then test for removable Po-210. Such experiments will be conducted in the near future.

The dirt layer on glass samples from dwellings constitutes extra weight that can sometimes be determined with precision scales. In particular, glass sheets covering diplomas, photos etc. can have a heavy load of dust and grease. Several dirty samples have been analysed for Po-210 before and after cleaning in this project and on no occasion have we been able to

Fig 15 Dark field analysis of a dirty glass surface. The dirt appears as a white pattern on a black background of clean glass. (magn. x500)

remove more than 50% of the Po-210 activity. This was also the case when the dirt layer corresponded to an even thickness several times greater than the recoil range. The explanation must be that the layer of dirt is unevenly distributed over the glass surface. In order to see if this was the case one of the dirty glass sheets was analysed in detail with a special light technique. The results are given in Figure 15. It is clear that even in this rather "dirty case" the greater part of the glass surface is open for recoil implantation. If a general conclusion can be drawn from the few pilot cases reported here it should be that loose contaminants on authentic household glass sheets interfere less with the alpha recoil implantation than initially thought or suspected. Systematic investigations in the future will tell if this conclusion is correct.

Rn/Po-210 correlation studies in homes

The Po-210/glass can not be "calibrated" by sampling glass sheets from dwellings for the simple reason that old radon levels can not be assessed in retrospect. One should bear this reservation in mind when investigating the correlation between implanted Po-210 and radon exposure in real dwellings. The correlation can never be better than the quality of the radon exposure estimates.

In this project a group of single-family houses in a village just outside Lund has been investigated. This group of houses was approached for several reasons. The main reason was that all the houses in the group are more or less identical and that only a few have undergone major reconstruction since they were built in 1967. The conformity of the dwellings is advantageous but as all the houses are privately owned, decisions concerning house repairs and energy-saving measures, for instance, have been taken on an individual basis. As the village is situated just a few minutes' drive from Lund the collection and delivery of glass samples for measurements in our lab was facilitated.

All dwellings investigated suffer from increased indoor radon levels due to the construction material, alum-shale lightweight concrete ('blue concrete'). The houses have no basement and the majority are self-draught ventilated with their own oil-fired heating system distributing hot water to radiators under the windows.

About 60 households were asked by letter if they were willing to participate in the study and 25 of them answered positively. These owners were asked to place a piece of glass from a photograph frame or similar objects at our disposal for a few days and in return they were offered a radon measurement free of charge. SSI supplied and evaluated the track-etch films⁽¹⁸⁾. The radon cup holding the CR-39 film strip is of the same type as that used by

Fig 16 *Distribution of bedroomfilms*

NRPB in Great Britain⁽¹⁹⁾. In each dwelling, one radon cup was placed in the room from which the piece of glass was taken and another cup was exposed in the bedroom. The exposure period covered two months, from March to April 1990. The distribution of the radon concentration values measured with the aid of the track-etch films are given in Figure 16.

The weather conditions were warmer than normal for southern Sweden during the track-film measurements. Very few nights were below the freezing point but the heating systems were in use throughout the major part of the film exposure period. Upon returning the radon cups the home owners answered a questionnaire about the age and location of the glass and more general questions about the construction, heating and ventilation of the house. Most of the pieces of glass supplied were analysed non-destructively in the open PIC. Usually, two or three glass samples were analysed in our laboratory per day.

Of the participating 25 dwellings, 22 supplied useful sheets of glass. For each of these 22 dwellings the accumulated radon exposure was estimated by multiplying the age (i.e. exposure time in the house) of the glass by the radon concentration value given by the single track-etch film exposed in the same room as the glass sheet. The measured Po-210 activity of the glass sheets was corrected for decay (old glass samples) or non-equilibrium with Pb-210 (young samples) and the corrected values were plotted against the estimated radon exposure

Fig 17 The Po-210 implanted activities of the 22 glass samples of the Po-210/Rn correlation project, corrected for decay and lack of equilibrium.

(Figure 17). The uncertainty in the individual CR-39 film values depends on the number of tracks counted. This statistical error is insignificant in Figure 17, corresponding to a relative standard deviation of less than 1% in most cases. The relative standard deviation from counting statistics in analysing Po-210 is, in most cases, about 5 %.

The assumption that the measured radon level has persisted for the whole exposure period, up to 24 years in Figure 17, can lead to a substantial error in some cases. Also, the exposure history of a sheet of glass can be more or less accurate depending on how well the home owner remembers, or can trace by notations, the history of that very piece of glass. Realistically, large errors must be associated with the accumulated radon exposure, and due to the involvement of human authenticity outliers cannot be excluded. Considering the uncertainty in the exposure estimate, based on a single track film value, and the variability in the plate-out phenomenon, the spread in Figure 17 is not surprising. There is a tendency for old glass samples to show a lower Po-210 value than expected from the linear trend. This trend can perhaps be explained by the fact that the radon levels several years ago were generally lower than today, due to less ambitious energy-saving measures. The coefficient of correlation (r^2) is equal to 0.54 in Figure 17.

CONCLUSIONS

The pilot studies performed show that long-lived radon daughters (LRnD) implanted into indoor surfaces can be used for retrospective radon monitoring. Due to availability, cleanness, and freedom from electrostatic influences, plane glass sheets openly exposed to radon and short-lived radon daughters (SRnD) seem to be the optimum choice for investigations of deposited and implanted LRnD. Background problems are significantly reduced and a high degree of specificity for airborne radon is achieved by alpha spectrometric analysis of implanted Po-210. An open-flow pulse ionization chamber has been developed and successfully utilized as a reference spectrometer for non-destructive investigations of large glass objects. Surface barrier detectors (SBD) and the like are less sensitive than the pulse ionization chamber due to the much smaller effective area, but are more versatile. An SBD detector can be used for *in situ* measurements of Po-210 under conditions of constant temperature.

As the system is not effected by electrostatic forces, the SRnD plate-out pattern is homogeneous on a cm. scale, but on a larger scale several non-dose-correlated factors may cause variations. Windows with radiators beneath, for instance, show a tendency towards increased plate-out compared with glass surfaces hanging on indoor walls. Improved air mixing during the winter season is thought to be responsible for the enhancement, but thermophoretic forces may also play a part. Contaminants residing on indoor glass surfaces may catch the alpha recoils, but the results of this investigation indicate that dirt particles influence the glass implantation process only to a minor extent, and at any rate less than hitherto assumed.

Single glass samples from 22 dwellings were analysed for implanted Po-210 and the levels correlated to the contemporary radon level. Correcting for non-equilibrium or decay, the Po-210 values correlated well ($r=0.73$) with the estimated radon exposure, considering the variation in plate-out and the uncertainty in the radon estimate.

Clearly, the practical exploitation of Po-210 in glass as a retrospective radon monitor must proceed with great care as the effects of modifying and non-dose related factors under realistic indoor conditions are poorly known. Pending the improvement of knowledge in this field it is advisable to analyse more than one glass sample of well known exposure history from each dwelling investigated.

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APPENDIX A

EQUIPMENT USED FOR THE CLOSED IONIZATION CHAMBER

Detector:	Cylindrical vessel (made in collaboration with an Australian laboratory, OSS, Jabiru, NT) volume 30 l
Material:	Stainless steel, inside painted with a distemper (Araldite)
Preamplifier:	Canberra 2002
Amplifier:	Tennelec TC 205A
High-voltage supply:	Grid: Tennelec TC 948 Anode: Tennelec TC 950
Pressure gauge:	Displaying: -100 kPa to 0 kPa
Gas filling:	P-10 (10% methane + 90% argon); the main contamination is nitrogen and the gas has a purity of 99.995%.
Drying Agent:	P ₂ O ₅ (diphosphorpentoxide)

Detector Data and Settings:

Cathode diameter:	19 cm
Grid diameter:	23.5 cm
Grid-wire diameter:	0.13 mm
Grid-wire step:	1.55 mm
Anode diameter:	21.8 cm
Cathode-grid distance: (flexible distance possible)	122 mm
Grid-anode distance: (flexible distance possible)	28 mm
Inefficiency of the grid (calculated)	0.011
Voltage:	cathode: 0 V grid: 1500 V anode: 3000
Operating gas pressure:	atmospheric
Energy resolution (FWHM):	120 keV (^{244}Cm); 72 keV (^{210}Po glass)
Detection efficiency at full source size:	0.65
Background (at 200 keV window at 5.8 MeV):	$6.5 \cdot 10^{-3}$ cps
Shaping Time Constant:	8 s
Coarse Gain:	20
Fine Gain:	1095

APPENDIX B

EQUIPMENT USED FOR THE OPEN IONIZATION CHAMBER

Detector:	Cylinder (made at our own laboratory); volume 6.8 l
Material:	polypropene, painted with silver paint
Preamplifier:	Canberra 2002
Amplifier:	Tennelec TC 205A
High-voltage supply:	Grid: Canberra RI 0113 Anode: Tennelec TC 945
Pressure gauge:	Platon (displaying :5 ⁰⁰ cm ³ /min -10 ⁰⁰ cm ³ /min air) Kyrölä
Pressure gauge:	Dwyer
Flow Rate Display:	BS 100
Gas filling:	P-10 (10% methane + 90% argon); N45

Detector Data and Settings:

Cathode diameter:	19 cm
Grid diameter:	16.5 cm
Grid-wire diameter:	0.16 mm
Grid-wire step:	2.0 mm
Anode diameter:	16.5 cm
Cathode-grid distance:	65 mm
(distance can be varied in discrete steps of 1 cm)	
Grid-anode distance:	35 mm
Inefficiency of the grid:	0.039 (calculated)
Voltage:	cathode: 0 V
	grid: 600 V
	anode: 1500 V
Operating gas pressure:	slight overpressure
Energy resolution (FWHM):	190 keV (^{244}Cm), 63 keV (^{210}Po glass)
Detection efficiency at full source size:	0.53 0.65 (collimated)
Background (at 200 keV window at 5.8 MeV):	1.0 10^{-2} cps
Shaping Time Constant:	8 s
Coarse Gain:	4
Fine Gain:	650
Pressure:	+20 to +200 Pa
Flow Rate Display:	2^{∞} - 3^{∞} cm^3/min (1 cm^3/min air corresponds to 0.771 cm^3/min of P-10 gas) (1 cm^3/min argon corresponds to 1.064 cm^3/min of P-10 gas)

APPENDIX C

EQUIPMENT USED FOR THE SURFACE BARRIER DETECTOR

Detector:	EG&G ORTEC BR-090-4000-100-S; with a chamber made at our laboratory, material is aluminium.
Preamplifier:	Canberra 2003 (resistance $R=110\text{M}\Omega$)
Amplifier:	Tennelec TC 205A
High-voltage:	CMTE 0-6 kV (with the possibility of reading both current supply and voltage).

Detector data, specifications and settings

Active Area:	4000 mm ²
Sensitive Depth (minimum)	100 μ m
Electrode Thickness:	50.0 gm/cm ² (Al)
Operating Bias:	- 100 V
Reverse Current (max.):	1.35 A (at 20 °C)
Temperature:	25°C
Shaping Time Constant:	0.5 s
Voltage:	- 212 V (-100 V to the detector)
Alpha Resolution: FWHM for ²⁴¹ Am (given by the manufacturer)	72.3 keV
Noise Width:	60.8 keV (FWHM pulsegenerator)
Coarse Gain:	200
Fine Gain:	815

A 1.5 μ m thick mylar foil was placed in front of the detector to protect it from contamination.

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