GLASS AS A RETROSPECTIVE RADON DETECTOR

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ABSTRACT

The combination of vitreous glass as substrate, alpha-emitting Po-210 as tracer, and a pulse ionization chamber as detector is a very promising retrospective radon exposure system for dwellings. The difficulties encountered at the moment are mainly concerned with the problem of finding glass sheets with well known radon exposure histories in order to test the accuracy of the method. Results from sheets of glass taken from six different houses are reported. The Po-210 activity P (Bq m^{-2}) of the glass surface correlates well with the cumulated radon concentration C (kBq y m^{-3}) estimated from long-term radon or radon daughter measurements. The correlation coefficient is 0.96 and linear regression analysis gives as best fit (+/- 1 S.D.) the equation $P = (0.91 \pm 0.11)C + (0.8 \pm 1.2)$. As a retrospective radon system the presented glass-polonium method has the potential to "see" several decades back in time, as well as being an excellent and truly long-term perspective radon detector.

INTRODUCTION

In any epidemiological study an appropriate and exact 'exposure' measure is desirable. In the case of radon the long delay between radon exposure and the manifestation of lung cancer is a serious problem. A radon or radon daughter detector that could look backwards in time would be ideal. With such a retrospective detector we should be able to determine the exposure relevant for today's cancer cases.

A retrospective radon system can be based on two different principles. Either you look for the radiation damage left behind after the decays or you try to detect the faint activity of the long-lived radon daughter Pb-210 or its decay products. In order to be of general use the substrate for the radiation damage or the material containing the Pb-210 activity must be present in all dwellings. Alpha track damage detectors fail in this respect as, unfortunately, the tracks can be made visible only in certain plastics not found in all dwellings. For a retrospective radon system we are thus left with the problem of finding if a home has a 'memory' of its cumulated radon concentration via Pb-210 (or its daughters) and if this 'memory' is strong and reliable enough. Just recently, our radon group in Lund showed that the activity of Po-210 (a daughter product of Pb-210) in glass sheets from dwellings was detectable and highly correlated to the cumulated radon exposure (1). In the rest of my talk I will explain the principles underlying this retrospective radon method and report our latest results.

FUNDAMENTALS

In Figure 1 the decay scheme of radon is shown to the left, and the right-hand part illustrates, very schematically, the principles behind the method. Some short-lived daughters are of course ventilated away, some decay in the air etc., but the important pathway to us is through the deposition of some daughters on macroscopic surfaces. This adsorbed activity is only loosely attached to hard surfaces, but there is a way in which the activity can be permanantly caught. The definite penetration into a surface, subsequent to an alpha decay, is a plain and simple recoil process already described by Hahn and Meitner in 1909 (2). In 50% of the alpha decays the alpha particle is emitted outwards and the daughter nucleus inwards. The maximum range in glass of the recoiling nucleus is about 0.1 micrometres, deep enough to save the recoiling atom from being scavenged even by the most elaborate household cleaning practices.

In hard materials, such as stone and glass, migration of large non-gaseous atoms is a very slow process. That is to say that during constant radon and room conditions we have a constantly increasing number of Pb-210 atoms absorbed superficially and permanently in hard materials.

Figure 2 illustrates the increase in absorbed Pb-210 activity over a 50-year period. The growth curve displayed is the result of theoretical calculations with a compartment model using typical rate constants for the indoor environment. We conclude from this slide that during idealized conditions the 'Pb-210 memory' is useful over several decades, but also, in order to render true linearity, we have to make corrections for radioactive decay of Pb-210 for radon exposure times in excess of about 10 years.

DETECTABILITY

Again using reasonable rate constants for the indoor situation we can predict the Pb-210 activity absorbed on hard surfaces as a function of cumulated radon exposure. Such order-of-magnitude calculations reveal that following an exposure of 1 kBq y m^{-3} radon-222 (27 pCi y l^{-1}) we can expect a surface activity of Pb-210 not too far from the range 1-10 Bq m^{-2} (2.5-25 pCi ft^{-2}). A surface activity of Pb-210 with such low intensity, the lower limit corresponds to approximately one decay per cm^2 every third hour, is indeed difficult, if not impossible, to resolve from the background in beta and gamma spectrometers. (In 4% of its decays Pb-210 emits a gamma quantum of energy 47 keV).

To be able to disclose 1 Bq m^{-2} it is better to turn to Po-210, the alpha-emitting granddaughter of Pb-210, and to large-area alpha spectrometers. A pulse ionisation chamber can analyse the 5.3 MeV alpha from Po-210 over a surface of several hundreds of square centimetres and such a detector was our choice when we started to look for alpha activities on glass sheets about a year ago.

The alpha spectrometric method has the advantage of being radionuclide specific and, in addition, it is easy to distinguish between the superficial activity we are looking for and alpha activity distributed homogeneously in the glass body. The only disadvantage of using Po-210 instead of Pb-210 is that the activity of Po-210, according to theory, is lower than that of Pb-210 and more dependent on radon exposure time. The expected activity ratio Po-210/Pb-210 in glass as a function of radon exposure duration is shown in Figure 3.

RESULTS AND DISCUSSION

So far we have analysed glass sheets from six dwellings, four in Sweden and two in Finland. The relation between Po-210 activity absorbed on the glass surface and the cumulated radon concentration is given in Figure 4. All eight glass surfaces showed easily detectable amounts of

Po-210, due to the high sensitivity of the detector. Glass samples from houses 5 and 6 were small enough to fit into the detector directly. All other sheets were cut to an area of about 210 cm^2. In houses where only radon daughter measurements have been performed, we used an equilibrium factor of F=0.5 to convert to radon concentration.

In Figure 4 the glass sheet from house No. 6 seems to be an outlier yielding too low a Po-210 value. This glass sheet is different from the others as it is slightly etched on the exposed side. In our opinion, however, this cannot explain the low polonium activity and in order to get more information than just the cumulated radon concentration value we contacted Dr R. Mustonen who sent us the sample. He confirmed that the house in which the glass sheet had been exposed had very well documented radon levels and that the estimated exposure, 17.8 kBq y m^{-3}, must be quite close to the truth, but also that the frame holding the glass had been standing in a bookshelf. Our hypothesis is now that the stationary air in the bookshelf gives rise to a lower plate-out rate than in the open, where the radon daughter measurements are taken. This phenomenon may be wholly responsible for the low Po-210 value for the glass sample from house No. 6. Awaiting new glass samples from the house the value in Figure 4 is considered as an outlier and excluded from the regression analysis.

That the glass sheet must be openly exposed to the room aerosol in order to be useful is supported by our results from one of the glass sheets from house No. 2. For this cupboard glass the Po-210 activity on the side facing into the cupboard is only 52% of the activity on the outward facing side given in Figure 4. In Figure 4 we also notice that the Po-210 activities on the two sides of the door glass from house No. 4 are different, indicating that the radon levels or plate-out intensities have been different in the two rooms separated by the door. Unfortunately, this hypothesis could not be tested as countermeasures against radon have recently been taken in the house.

Applying linear regression analysis to the data in Figure 4 (excluding data from house No. 6) yields an intercept not significantly different from zero, 0.9 ± 1.2 Bq m^{-2} and a slope of 0.91 ± 0.11 m ky^{-1}. The error interval given is 1 S.D. The correlation coefficient has the value 0.96. Considering the difficulties in obtaining the correct radon exposure values for the glass sheets and the complexity of radon aerosol chemistry and plate-out, the spread in Figure 4 is astonishingly small.

The "well behaved" data in Figure 4 is presumely due to several factors:

1) Long-term radon measurements performed by reputable laboratories.

2) Carefully selected houses with no or small building and ventilation alterations during the exposure time. (The glass sheet exposed for 25 years in house No. 2 is an exception to this and in house No. 1 the radon concentration was remeasured after the installation of a new ventilation system.)
3) Large surface and low electrostatic charging of the substrate glass.
4) Most glass sheets are true indoor ones. On windows the variations are probably larger.
5) A simple physical effect (nucleus recoil) is responsible for the absorption of Po-210.

The correlation in Figure 4 could have been even better if we had known the radon exposure of the glass more accurately. Only in one house (No. 3) has the radon measurement been taken close to the position of the glass sheet.

The sides protected from room air show no, or at least very little, traces of Po-210. This fact is illustrated in Figure 5. The narrowness of the alpha peak proves that the Po-210 activity is confined to the outermost layer of the glass.

CONCLUSIONS

The presented glass-polonium method constitutes a breakthrough in the ability to measure radon in dwellings retrospectively and has put a long-term radon detector in all houses that have glass windows or other glass sheets. The method is not restricted to glass, in principle any hard non-electrostatic material with a flat, large surface area that is openly exposed to the short-lived radon daughters will do provided the frequency of cleaning is less than say once per day. With a pulse ionizing chamber as a detector for the Po-210 activity, the sensitivity is high enough for the method to be used even in houses with with low radon levels, provided the exposure time is long enough. Today the experimental results are too scarce to validate any final judgement of the usefulness of the method but the data so far are very promising. Future investigations should be focussed on the inherent variability in the method caused by variations in radon daughter plate-out and other phenomena influencing the transfer of activity from radon-222 in the room air to the absorbed Po-210 in hard materials. The best way to 'calibrate' the method is by the investigation of glass samples exposed to radon authentically in dwellings under well-controlled conditions.

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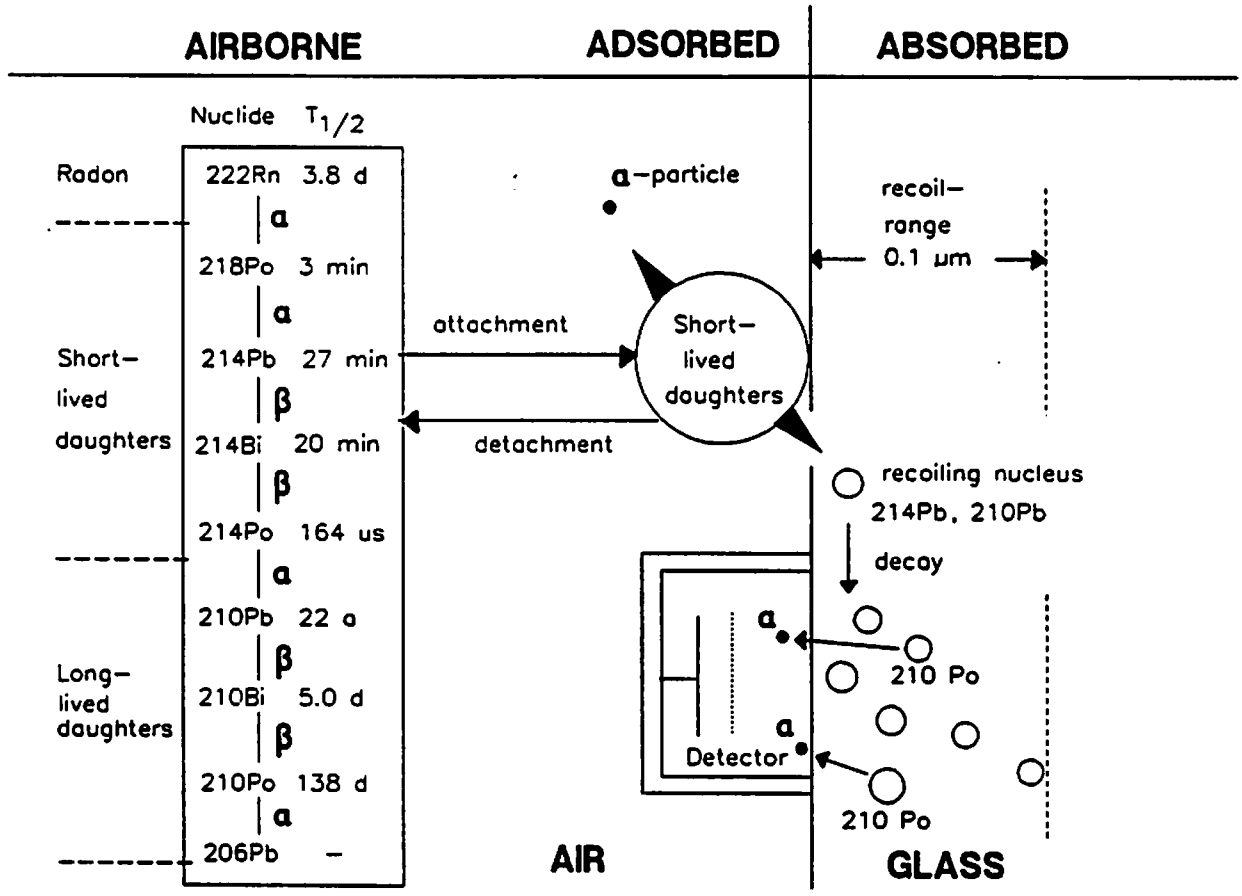

Figure 1. A schematic illustration of how the short-lived radon daughters are attached to a macroscopic surface (glass) by plate-out and how the subsequent alpha decays permanately embed the daughter nucleus in the outermost layer of the glass by a recoil effect. From measurements of the Po-210 activity of the glass using alpha spectrometry, the accumulated radon concentration can be determined.

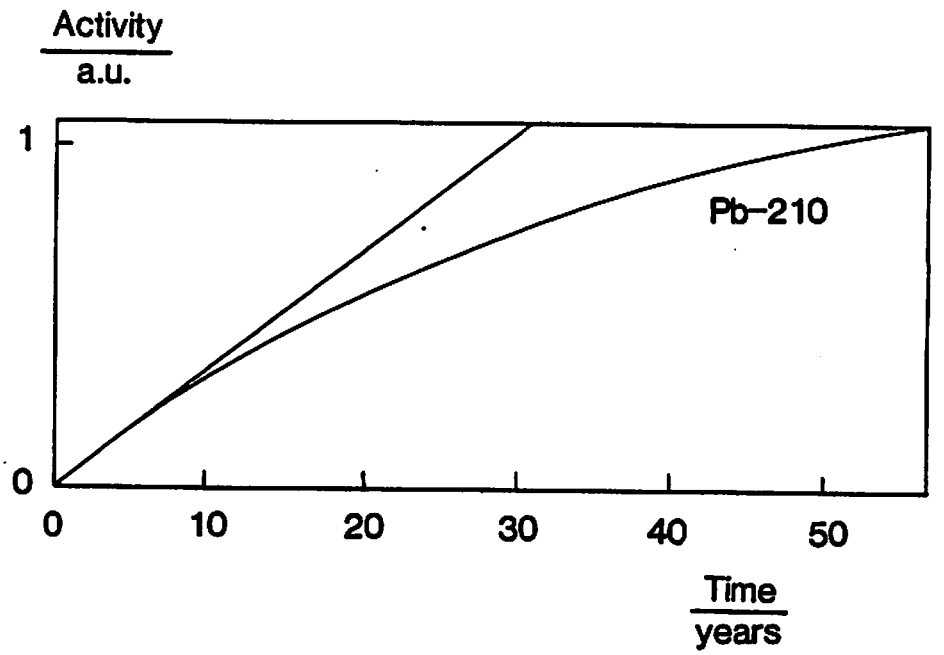

Figure 2. The growth curve of Pb-210 activity absorbed in a hard material due to alpha recoil effects as a function of radon exposure time, assuming a constant radon source is opened at time zero (theory). The initial slope of the curve is also shown.

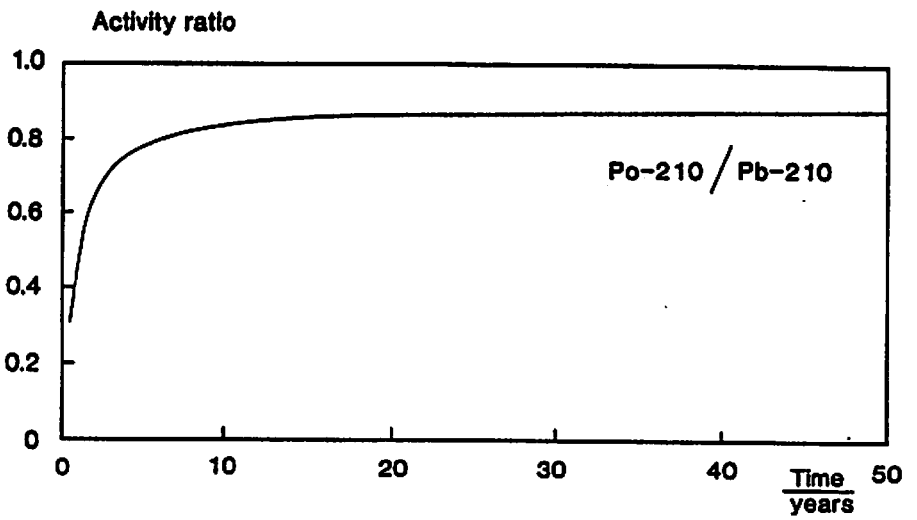

Figure 3. The same as in Figure 2, but showing the activity ratio between Po-210 and Pb-210.

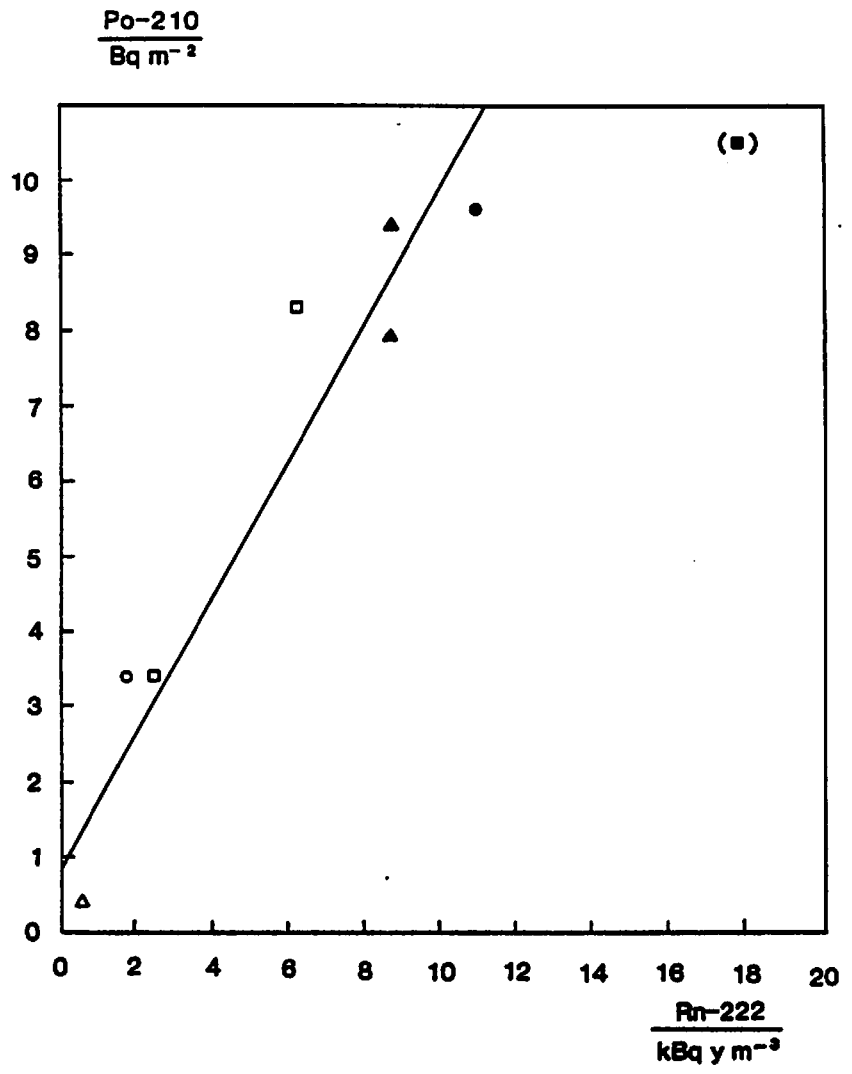

Figure 4. The Po-210 activity in glass per unit area as a function of the estimated integrated Rn-222 activity concentration. The linear regression line excluding house No. 6 is shown (see text). For the interior door glass (house No. 4) the activity on both sides of the glass is plotted. (1 Bq = 27 pCi, 1 kBq y m^{-3} = 27 pCi y l^{-1}).

Legend:	△	House: 1	Exp.time: 17y	Glass: photograph
	□	House: 2	Exp.time: 10y,25y	Glass: cupboard,window
	○	House: 3	Exp.time: 10y	Glass: picture
	▲	House: 4	Exp.time: 14y	Glass: door
	●	House: 5	Exp.time: 5y	Glass: photograph
	■	House: 6	Exp.time: 4y	Glass: photograph

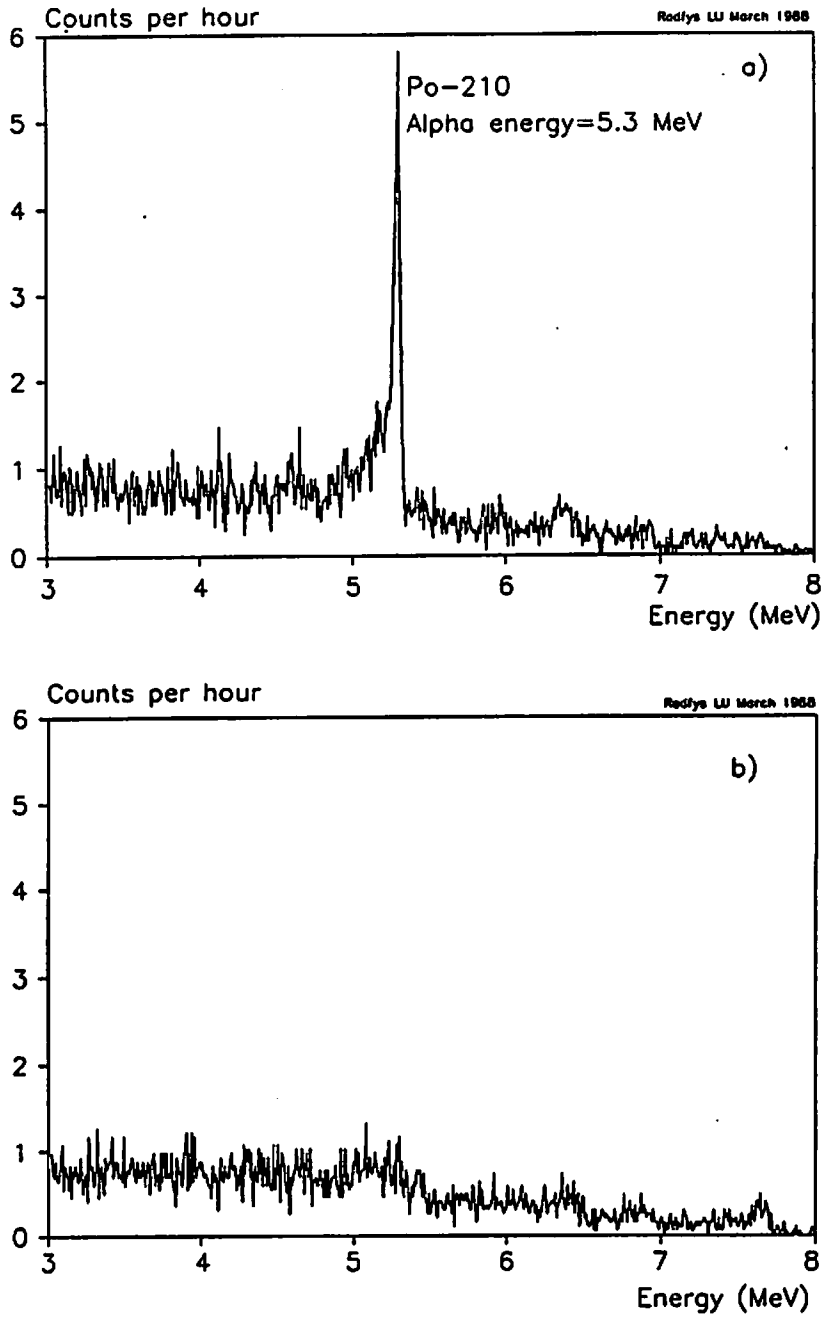

Figure 5. Alpha spectra from the glass sheet (the least exposed sample) from house No. 1, obtained in a pulse ionization chamber.

a. Side facing the room.

b. Side facing the photograph.