

Radon Progeny Recoil Effect in Retrospective Indoor Glass Dosimetry

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ABSTRACT

Radon gas diffusion and progeny transport in air, are mechanisms to be considered in retrospective glass dosimetry. With the aim to contribute to the understanding of the Rn progeny recoil energy role in this dosimetry methodology, we carried out a simulation employing GEANT4 code. In that, we assumed the chemical compound of the glass that is used commonly in households. Results are compared to experimentally measured ²¹⁰Pb concentration to show that the recoil energy helps the progenies incrustation, mainly for the ^{218,214}Po alpha emitters but do not influence bismuth-210 diffusion directly. A significant difference exists between our results and measured values; that is interpreted as due to atomic displacement by primary knock-on atoms. The SiO₂ molecule binding energy breaks and the following ion recombination, induce a structural modification between the atom by e.g. cavities formation in such a way that reduces significantly the radon progeny diffusion speed..

1. Introduction

Low level radon exposure during long period of time has its importance since exposure to the radioactive gas and progenies may be linked to lung cancer and childhood leukemia. The evidence of alpha emitter nuclides that induce neoplasia is suggested by studies of miners and laboratory experiments [1]. In this work we study one method of measuring such exposure, namely retrospective indoor glass dosimetry RD.

During decay of radon-222 its short-lived progeny RnD, can plate-out onto the surrounding matter; for instance, alpha-recoiled radionuclides can get encrusted in glass. A scheme is given in Fig 1 to represent the multi-step displacement process through surface glass that relies on the alpha recoil energy of binary radionuclide ²¹⁸Po – ²¹⁴Pb double, or ²¹⁴Po – ²¹⁰Pb simple implantation. The large radionuclide Coulomb field displace the smaller glass atom gaining new position until the next decay provides kinetic energy to move again. However, it is expected that the transport by diffusion is the primary mechanism to penetrate deeper the glass surface.

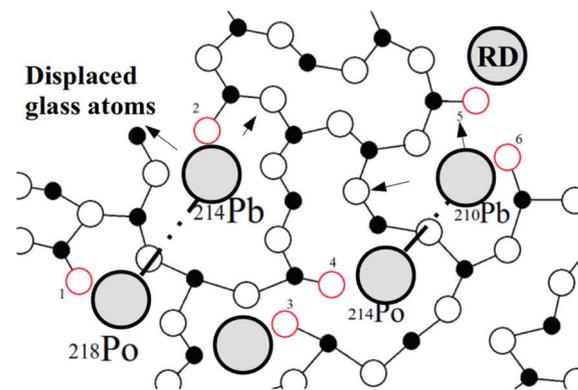


Figure 1: Artistic view of surface implanted RnD radionuclide penetration in the glass driven by recoil energy.

The concentration of implanted nuclides, such as bismuth, can provide the necessary information to determine the levels of past indoor radon activity and exposure to humans. The most important decay chain of ²²²Rn are reported in Figure 2 (some radionuclides such as the astatine, thallium, mercury has a negligible occurrence and it is not included in the study).

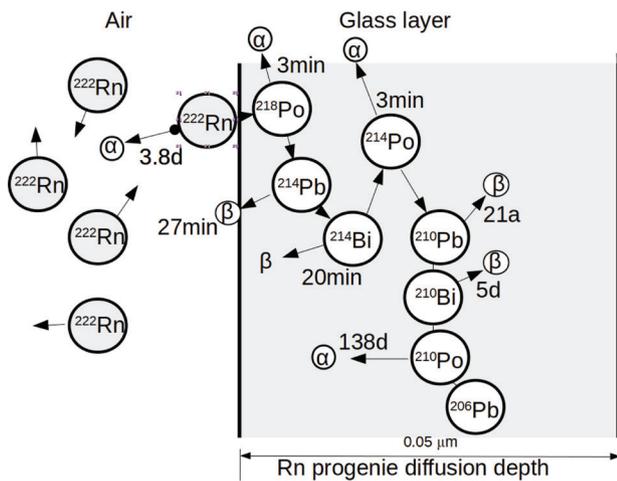


Figure 2: Artistic illustration of Rn-progenies ^{218}Po diffusion in household glass. The mechanism imply that every radionuclide has a given probability to move deeper in the superficial glass. It is expected that recoil energy provides a boost to furthering diffusion inside the matter. The illustration shows only selected radon progeny.

The implantation of naturally occurring radionuclide depends on whether the recoil energy of alpha emitters progeny is sufficient to penetrate the superficial layers of glass. Crooks reported in 1903 that radium daughter recoil energy of 84 keV is sufficient to detect a fraction of them below several atomic layers, a depth at which ^{210}Pb is retained [2]. Measuring the concentration ^{210}Bi , a naturally implanted radon progeny, in any subsurface layer, can provide time-integrated indoor radon concentration; a method developed for retrospective dosimetry (RD) employing glass, ceramic, or enameled objects as reported by Mahaffey [3] and determined experimentally by Pálfalvi [4] in 1995. A year later Falk [5] determined that RD by passive nuclear track methodology relying on the autoradiography, provide exposure rate around $1\text{ kBq}\cdot\text{y}\cdot\text{m}^{-3}$. More recently, Martin et al. [6] applied the method in epidemiological studies employing mirror glass surfaces. Figure 2 shows a double implantation scheme from ^{218}Po , and single implantation from ^{214}Po , taken from Roos [7].

These RD methods rely on measurements of the superficial activity of a glass object, and work thanks to the half-life of 22.26 y of ^{210}Pb long, as reported by Lively and Ney [8] and later by Samuelsson [9]. The reconstruction mechanisms of past indoor radon concentration assume that over decades all the radon progeny stored in the glass, such as ^{210}Bi , is produced by airborne radioactive gas. The advantage of the technique is that it can be applied in dwellings that have been occupied and exposed for long times; cases in which the indoor dose can be determined by measuring the activity on the inner surface of glass windows, for instance. Results given by an epidemiologic study suggested that the

glass-based methodology is more accurate in comparison to few other, when radon exposure related to lung cancer risk has to be estimated, (Alavanja et al. [10]). Lagarde et al. [11] report that glass-based long term dose estimates, provide a more relevant exposure proxy in comparison to air-based estimates.

The aim of this study is to use the program Geant4 to simulate the absorption of the radon progenies in glass as well as its spatial distribution and compare these results with experimentally observed retrospective dosimetry.

2. Simulation of Radon Progeny Diffusion in Glass

Geant is the name of a computer program created from “Geometry”, “And” and “Tracking” used to simulate the interaction of particles with a media, e.g. a beam with a target in particle physics experiments. In our case we use GEANT4, the 4th version of the computer code, to simulate the interaction of Rn progenies with a glass. The simulation is composed of the following steps:

1. The geometry of the glass medium is defined.
2. The progenies of ^{222}Rn and their kinetic energies are specified.
3. The processes each ^{222}Rn daughter can undergo are specified.
4. An Rn progeny (α , ^{218}Po) is created in a given direction and with certain kinetic energy.
5. The α and ^{218}Po are transported through the glass starting a chain of decays according to Fig. 1.
6. The data (particles produced, location, energy) from each event are stored.
7. A new event is generated as in step 4.

In the present application, the glass is in air and there are no other external factors (such as electric or magnetic fields, nor detector response issues). Ionizations that induce secondary electrons were not taken into account due to the complexity of associating them with the diffusion phenomenon. The geometry used was a rectangular volume of 1 mm of thickness, and an area of 20 cm x 20 cm. Figure 3 shows the geometrical setup and Figure 4 a typical set of GEANT4 events.

The ^{218}Po decays through alpha transitions with a half-life of 186 s and the simulation will allow us to know how deep it goes in the glass during an event (hardly an appreciable distance). Other decays product, such as ^{210}Pb , will be able to penetrate deeper into the glass due to their longer half-lives. The simulation follows a few hundred of Rn-progenies nuclide. The influence of the alpha recoil energy on the diffusion phenomenon has been determined by comparing the displacement of binary radionuclide of $^{218}\text{Po} - ^{214}\text{Pb}$, or $^{214}\text{Po} - ^{210}\text{Pb}$.

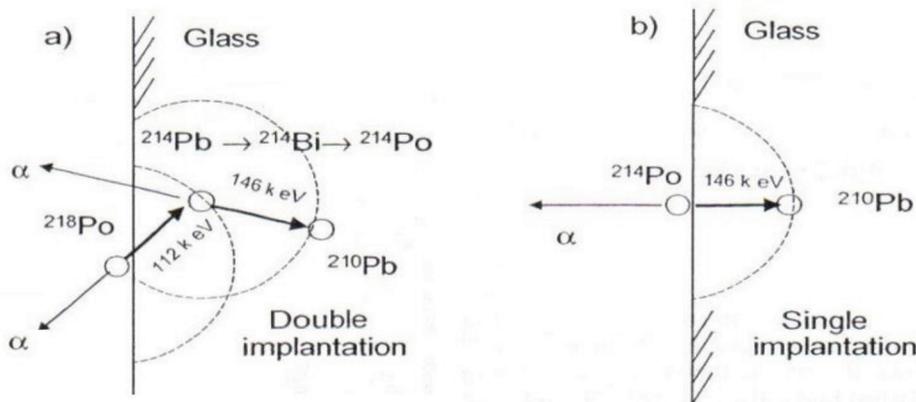


Figure 3: Double implantation from ^{218}Po (a), and single implantation from ^{214}Po (b), taken from Roos, 2002.

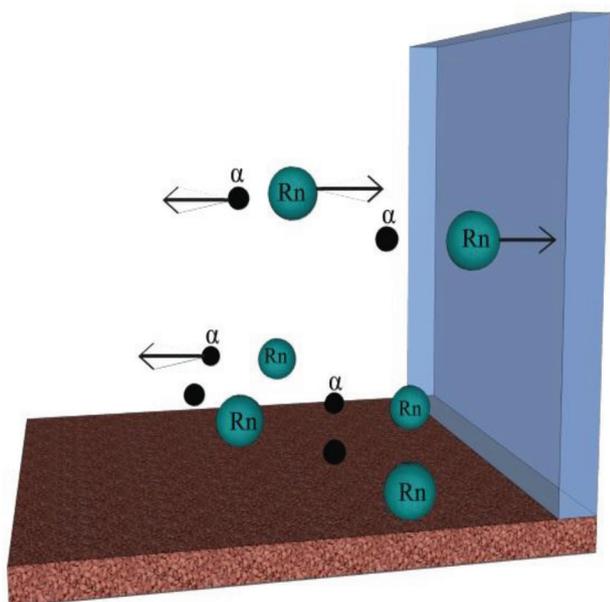


Figure 4: Geometrical setup showing the progeny of ^{222}Rn hitting glass (mainly SiO_2). Other radionuclides (Rn+Rn and helium) may be transported by diffusion or attached to suspended particular matter (spm) and may leave the space near the glass surface.

The material used in the GEANT4 simulation is window glass (specifically a glass with silica + sodium oxide (Na_2O) + lime (CaO) + alumina (Al_2O_3) + magnesia (MgO) [12] and other compounds with a concentration lower than 1%, listed in Table 1. The GEANT4 code produces data output that includes a list of the radioactive nuclei, their depth deposition, their activity and average free path. Of particular importance is the depth for the first two radon progenies, specially the activity of the radionuclide ^{210}Bi in the glass. The results of the code were compared with the experimental results and using the lists, given by [13], and binned in intervals of 5 years up to 50.

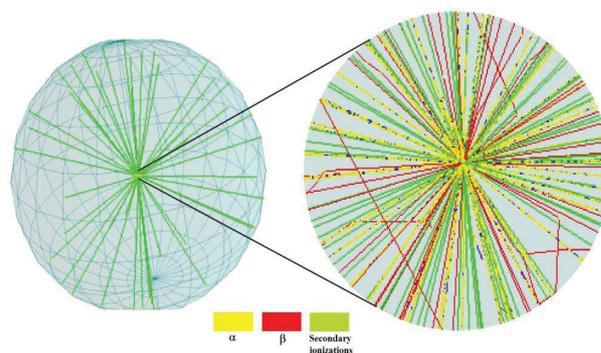


Figure 5: GEANT4 geometry used in the simulation of the plate-out of ^{222}Rn progenies. The sphere is the glass medium, and the different rays represent the α , β and secondary decays.

Table 1: Chemical composition of window glass with elements considered in the simulation with Geant4.

Chemical structure	% concentration in weight
SiO_2	74
Na_2O	13,1
CaO	10,5
Al_2O_3	1,3
K_2O	0,4
SO_3	0,3
MgO	0,25
Fe_2O_3	0,14
TiO_2	0,01
Sum	100

3. Diffusion Theory

To determine the implantation profiles of Rn progenies it is useful to study the diffusion equation through Fick's law. The diffusion mechanism in glass start at the surface with the first Rn-progeny; others follow as reported in Figure 1. The

diffusion phenomena is described by the one dimensional equation that is applied for a closed system and constant transversal cross section (Fick [14]):

$$\frac{d\Phi}{dt} = -D \frac{d^2\Phi}{d^2x} \quad (1)$$

where Φ [counts/m³ o mol/m³] is the concentration of bismuth, and D [m²/s] is the diffusion coefficient. Thickness of the glass and large exposure time provide a length and time scale to the problem, it is possible to use dimensionless units. The solution for equation (1) can be obtained using Fourier's method of separation of variables:

$$\Phi = \Phi_0 E + \sum_{n=0}^{\infty} \frac{2\Phi_0}{n\pi} \sin(n\pi E) \exp(-n^2\pi^2 Dt) \cos(n\pi x) \quad (2)$$

Where E is a small number that corresponds to the thickness of a given glass layer. We are interested at $x=0$ as this corresponds to the surface activity of bismuth and for which, the rate of convergence of the above infinite series depends on the small parameter E . Thus, care has to be taken when truncating the series while fitting it to the experimental data.

4. Results

The results of the simulation are shown in Figure 6. Results of the code binned in intervals of 5 years up to 50 show the implanted activity of ²¹⁰Pb in glass after exposure to ²²²⁺²²⁰Rn at a layer of depth of 0.018 μm.

Figure 7 shows the depth distribution of ²¹⁰Pb at time intervals of 10, 20, 30, 40 and 50 y, again with a depth layer of ~0.018 μm, and calculated with one million events in 10 year intervals until 50 years; we recall that the half-life of ²¹⁰Pb is 22.3 y.

Figure 8 shows the depth of the implanted radon progeny as a function of time. The ²¹⁰Pb is the nuclide of the Rn progeny that is best deposited in glass [2]; in this case there is evidence of statistical losses of a tenth of the initial exposure of ²²²Rn, these losses are observed in the transition from ²¹⁰Pb to ²¹⁰Bi. The fit was obtained applying eq. (2).

The results reported by Pálfalvi [4] will be used as a reference. Pálfalvi determined the activity of ²¹⁰Bi on glass irradiated by ²²²Rn during 55 years, using by chemical etching as well as passive detectors (CR-39™ and LR-115). Figure 9 shows a comparison of the simulations with the experimental activity of the implanted ²¹⁰Bi as a function of time.

The alpha back-scatter energy favors the diffusion mechanism only for the first progeny, that is, at the beginning of the diffusion mechanism, this is due again to the short half-life of the alpha-emitter progeny of the ²²²Rn, so the ²¹⁰Bi will remain a longer period of time in the same thickness of glass.

The ²¹⁰Bi is found in one of the deepest layers at, approximately, 0.0184 μm from the glass surface, which implies that Bi is at its maximum diffusion and it is its progeny what continues moving deeper, i.e. ²¹⁰Po until it reaches its stable isotope ²⁰⁶Pb. Bi cannot move deeper in the glass due to the binding energy of SiO₂, main component of conventional glass. If the recoil energy of an alpha is 84 keV, then if the binding energy of SiO₂ is 102 eV [15], it could produce about 750 ionizations/nuclide, which is not too large compared with the number of molecules per cm³, which is of the order of 10²³ atoms/cm³.

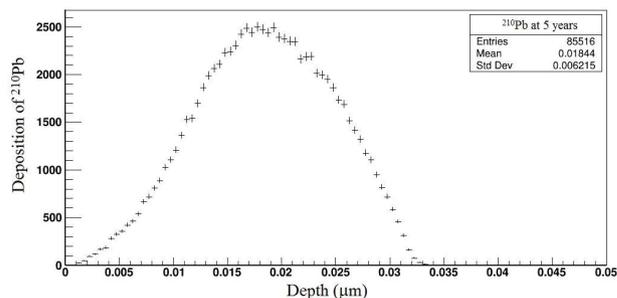


Figure 6: Deposition of ²¹⁰Pb in glass at five years after exposure to indoor ²²²⁺²²⁰Rn at a depth of approximately 0.018 μm.

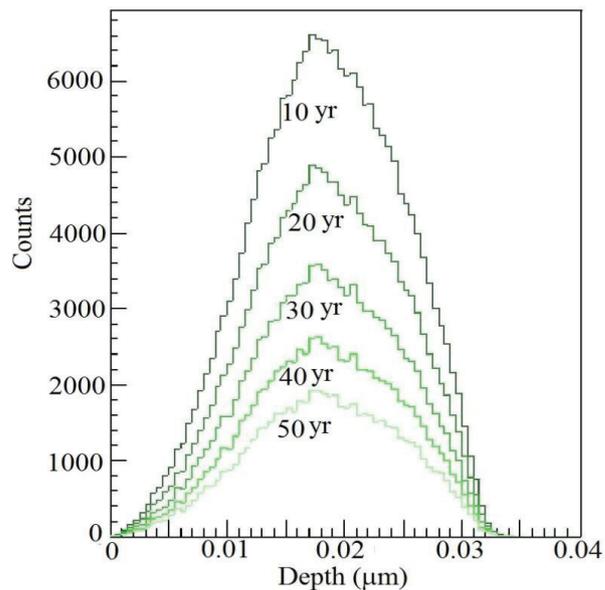


Figure 7: ²¹⁰Pb distribution with time interval of 10, 20, 30, 40 and 50 y, at ~0.018 [μm] depth layer.

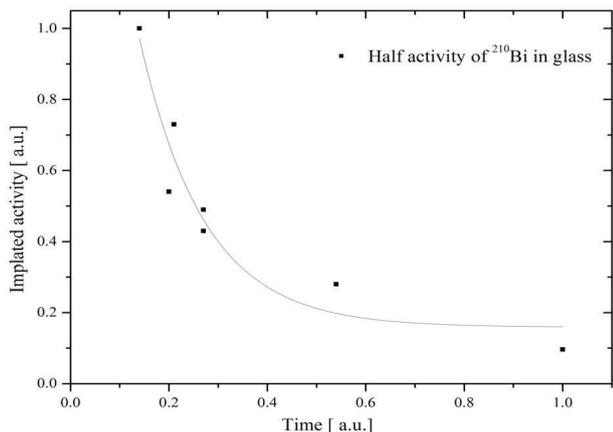


Figure 8: Surface activity. Ordinate indicate the radon progeny implanted depth as a function of elapsed time.

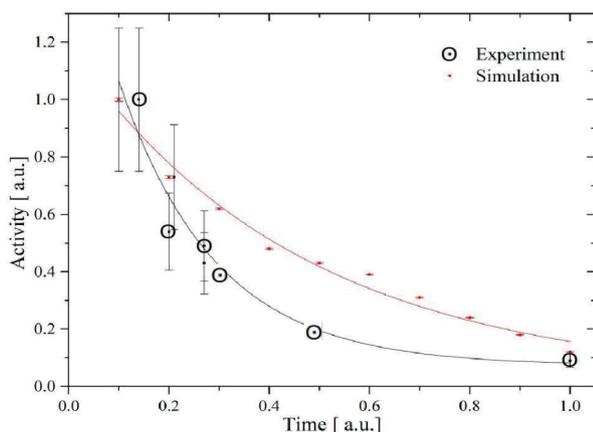


Figure 9: Comparison of experimental values reported by Palfalvi et al. With the results obtained with the simulation of the radioactivity of ^{210}Bi in glass, as a function of time in arbitrary units.

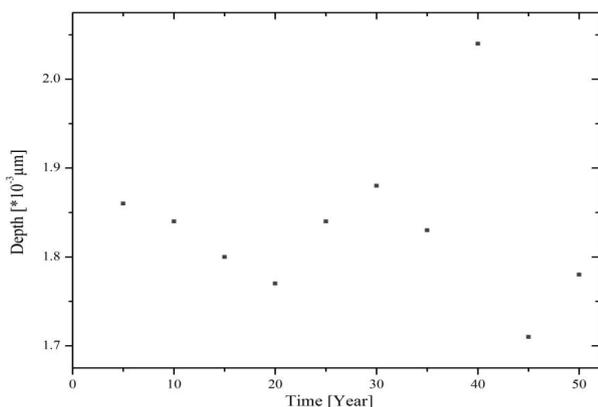


Figure 10: Average deposition distribution of ^{210}Bi in glass between 5-50 y the margin of error of the simulation is 0.006%.

Discussion

^{210}Bi is at a depth of approximately $0.0184\ \mu\text{m}$ in the glass, this means that it is the maximum diffusion length and it is its progeny that go deeper, the ^{210}Po , at the end it reaches, the stable element ^{206}Pb . The ^{210}Bi cannot penetrate further because of the bonding energy of SiO_2 . For conventional glass, as mentioned above, is $102\ \text{eV}$, while the alpha particle generates a recoil of $84\ \text{keV}$, corresponding to 750 ionizations/nuclide; at such a small energy few Primary Knock-on Atom or PKA will be displaced from its lattice site by the impacting heavy nuclei.

As shown in Figure 5, the ionizations presented by GEANT4 correspond mainly to secondary ionizations (green), and beta (red) and alpha (yellow) decays of ^{222}Rn are less in number. Therefore, although the secondary ionizations produce vibrations of the atomic net, it does not break any bindings nor causes a chain reaction that would propel the progenies deeper into the glass; the incident ions are thus fixed.

The simulation indicates that the volume of displaced ionized atoms (without taking into account the temperature) is of $257,109/300,000$ events. This is due to the high half-life of ^{210}Pb , and although the alpha-emitter nuclides of smaller half-lives get deposited and eventually decay, their progenies products contribute to a cascade effect.

After it is displaced from its initial site, the PKA can induce the subsequent displacements of other atoms in the case of carrying sufficient energy, otherwise come to a rest at an interstitial site where it stops until decay products start a new interaction [16]. It is well known that the main processes that contribute to the energy loss of a moving ion are the elastic interactions between the nucleus of the ion, and the Coulombic interactions between electron-screened nuclei and the inelastic interactions between the ion and the electron orbitals of the target atoms. Relatively small contributions to dE/dx may also arise from electron-transfer processes occurring between the moving ion and neighboring atoms. The nuclear and electronic contributions are not independent of one another because energy is transferred by both mechanisms during a single close collision.

As the ions are deposited in the glass, a loss of energy occurs due to elastic collisions between atoms and the ^{222}Rn progeny. By conservation of energy the progenies receive an impulse upon decay, such impulse is substantially larger in alpha decays than in beta decays. In 74% of the cases the loss of energy of the progenies of ^{222}Rn is manifested as high impedance and, as shown in Figure 5, making the ^{210}Pb get deposited abundantly near the surface.

Conclusions

Alpha back-scatter energy favors the diffusion mechanism of ^{222}Rn progenies mainly only for ^{210}Bi ; the rest of the members

of the family chain are less favored, except, perhaps, for the last one before reaching the stable isotope ^{206}Pb , which appears very slowly in the simulation.

The ^{222}Rn progenies with small half-lives, although present in the simulation, appears as an observable difficult to quantify for which its measurement is not recommended. A better observable is ^{210}Pb which, while being a daughter of ^{214}Po , the Rn progeny with largest alpha decay energy (7.7 MeV), is bound to yield an easier measurement.

In spite of observed differences between the simulation and experimental results of Figure 9, it is possible to agree that the model indeed simulates the implantation and diffusion of ^{222}Rn progenies satisfactorily with average deposition given in Figure 9. Most of the implanted ^{210}Pb does not leaves the medium in the time of the study, and the observed number of ionizations in SiO_2 (about ~ 0.85 /event) is reasonable and small compared to the number of atoms in the medium sample.

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