THE COMPACT DISC AS A RETROSPECTIVE RADON DETECTOR: PERFORMANCE OF THE METHODOLOGY

D. S. Pressyanov, Faculty of Physics
St. Kliment Ohridski University of Sofia, Bulgaria

ABSTRACT

Recently, a method for long-term ^{222}Rn measurements was proposed that combines the radon absorption ability of some polycarbonates, like Makrofol, with their track-etch properties. The method is independent of any particular ^{222}Rn progeny behavior and the very first evaluation of this approach showed that its range of sensitivity covers practically the entire range of indoor ^{222}Rn-concentrations. This method has been extensively studied in the period 2001 – 2007. In this report we comprehend the performance of the method as demonstrated in the laboratory studies.

INTRODUCTION

The indoor radon exposure is probably the second most important cause of lung cancer**,** after smoking [1]. Exposure for many years contributes to the risk, but up to now the duration of most of the integrated radon measurements is at maximum one year. In search of methods for representative long-term radon exposure estimates, considerable efforts were put in retrospective measurements. One of the approaches is based on the recoil-implanted ^{210}Pb/^{210}Po on glass surfaces [2, 3]. This "glass-implanted" method has a strong dependence on the particular behavior of radon progeny to aerosols in the indoor air and therefore its accuracy is at best a factor of two [4, 5]. Other approaches employ volume traps of spongy materials [6] or eyeglass lenses made out of CR-39 [7]. Methods based on volume traps give evidence of low sensitivity, low predictability and are strongly dependent on a suitable choice of spongy materials. Therefore they are of limited effectiveness for large-scale use. The eyeglass lenses method is affected also by a particular ^{222}Rn progeny behavior, and a non-uniform plate-out. Furthermore it was found that the CR-39 etching behavior could vary significantly from one supplier to another.

Recently, a method for long-term ^{222}Rn measurements was proposed that combines the radon absorption ability of some polycarbonates, like Makrofol, with their track-etch properties [8]. The method consists in the removal from the detectors, after exposure, of a surface layer with a thickness that is greater than the range of the most energetic alpha-particles coming from ^{222}Rn and ^{220}Rn progenies plate-out on the surface. After that, the detectors are etched electrochemically and the tracks are counted. Makrofol and equivalent plastics (e.g., Makrolon) are widely used as a basic constructive material for commercial CDs and DVDs. Therefore, home-stored CDs/DVDs may serve as retrospective long-term radon detectors. The method is independent of any particular ^{222}Rn progeny behavior and the very first evaluation of this approach showed that its range of sensitivity covers practically the entire range of indoor ^{222}Rn-concentrations [9].

Materials and Methods

In laboratory experiments, the CDs were exposed to controlled ^{222}Rn concentrations in hermetic laboratory vessels. The radon was supplied by certified sources. During the exposures the radon concentrations were followed by reference measurements. These measurements were made either with Lucas-cells, which were calibrated with a National Physical Laboratory (UK) primary radon standard, or by calibrated radon monitors (AlphaGUARD, Genitron Instruments GmbH). After exposure the detectors were left for more than 3 days (usually 2 weeks) to degas.

After exposure, a surface layer that is thicker than the range of the alpha-particles of the ^{222}Rn or ^{220}Rn progenies (i.e. ≥ 80 μm), is removed. The removal is made by chemical pre-etching (CPE). After that the detectors are electrochemically etched (ECE) and the tracks are counted. The tracks in depths ≥ 80 μm, are related only to the absorbed ^{222}Rn and any plate-out dependence is cancelled out. In most of the experiments the removed layer was about 80 μm, but in some of them the depth distribution of track density up to depth of 300 μm was studied. The CPE is performed at 25°C with an aqueous solution containing 45% KOH (m/v) and 40% methanol (v/v). Under these conditions the bulk-etch velocity is about 30 μm/h, and the target depth can be reached within a reasonable time.

After removal of the surface layer, the tracks are revealed by ECE for 3 hours [10] and counted automatically using a video-camera or computer scanner [11]. In the first experiments [9, 12] the etched pieces of CDs were of 300 μm thickness. The thickness of four spots of diameter 30 mm was reduced to 300 μm by mechanical treatment, by a lathe, from the back. Afterward, they were etched with 800 V (effective) at 2kHz. In 2006 a generator of higher voltage (up to 4 kV effective) and frequency of 6 kHz was constructed (Ekotronic Ltd., www.ekotronic.cz). With voltage of ≥ 2.5 kV (effective) and using the same ECE etching solution [10] the tracks can be revealed on CDs without need for mechanical reduction of their thickness. In the recent experiments in 2006 and 2007 we apply 3.3 kV effective. With such HV the etching efficiency for 1.3 mm thick CDs is similar to that in the initial mode of etching 300 μm detectors at 800 V.

RESULTS

Calibration experiments

Several experiments to study the correlation between the net track-density and integrated ^{222}Rn concentration have been conducted since 2001. In all of them almost perfect correlation has been obtained, as seen in Fig. 1. The CDs were exposed bare and in their jewel cases. In all experiments bare CDs and CDs in jewel cases gave the same results within the experimental uncertainty. Therefore, it was concluded that there is no effect of jewel cases. One of the benefits of the method is the possibility for *a posteriori*

calibration, e. g. after a CD is taken for analysis, to expose a piece of the studied CD to controlled integrated ^{222}Rn concentration and to use the data for individual calibration.

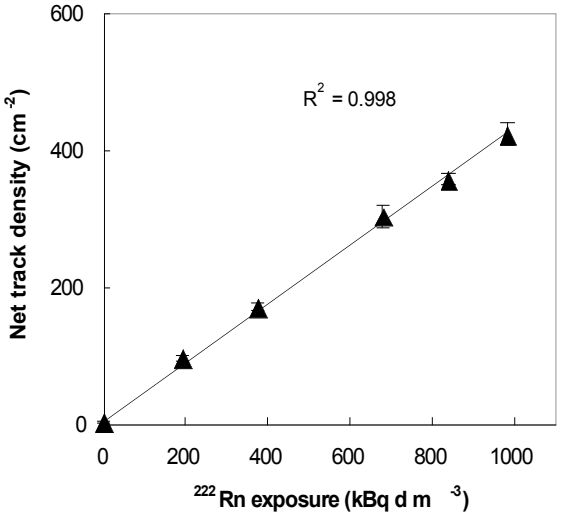

Figure 1. Correlation between net-track density and ^{222}Rn integrated concentration at 83 μm depth.

Experimental evidence of absence of contribution from the plate-out of the radon progeny

From the very beginning of the study of this method, attention was paid to whether the alphas from the radon progeny plate-out on the detector surface can contribute to the tracks at the studied depth. Experimental evidence that there is no plate out contribution has been comprehended in our previous works [12, 13]. Here we present one of the experiments that probably gives the most direct evidence for the absence of plate-out contribution. During this experiment 10 detectors were exposed to one and the same ^{222}Rn concentration. For half of them the 83 μm layer was removed mechanically. Thus all latent tracks from the α-particles coming from the detector surface have been completely removed. The remaining half was treated in the standard way, by CPE. Afterward, both sets were electrochemically etched using the standard procedure. The track density of the mechanically treated detectors was 297±16 cm^{-2} and that of the detectors treated by CPE was 289±22 cm^{-2}. The fact that the track density in both cases is statistically identical presents a direct evidence for the absence of any plate-out contribution to the tracks at the studied depth.

Influence of environmental factors

During the laboratory experiments the effect of the following environmental factors has been studied:

- Pressure;
- Humidity;
- Temperature;
- Cigarette smoke and dust deposition;
- Presence of ^{220}Rn.

The effect of all listed factors but the temperature is either missing or restricted within 10%. The last was observed under conditions that are unlikely to be common indoors, e. g. 100% of relative humidity or extremely dense cigarette smoke. The effects of pressure and humidity are illustrated in Fig. 2 and Fig. 3. More details of these experiments are given elsewhere [12, 13]. The effect of the temperature is more pronounced, as can be seen in Fig. 4. The temperature variation within $5^0 - 40^0$ C can introduce ±30% bias in the results. However, the results can be *a posteriori* corrected using the depth distribution of track density. As seen in Fig. 5 the depth distribution is close to exponential, the slope of the line (in log-linear scale) being dependent on the temperature. The idea for *a posteriori* temperature correction is by using the track density in two or more different depths to determine experimentally the slope and to use it to reconstruct the average temperature during exposure. As shown in Reference 12, if this correction is applied, the uncertainty in the long-term retrospective ^{222}Rn measurements by CDs could be potentially better than 10%. Theoretical estimates showed that the bias due to the growing ^{210}Pb+^{210}Po after long exposure times is about 8% for 20y exposure [12]. This bias may be corrected by applying a factor that depends on the time of exposure.

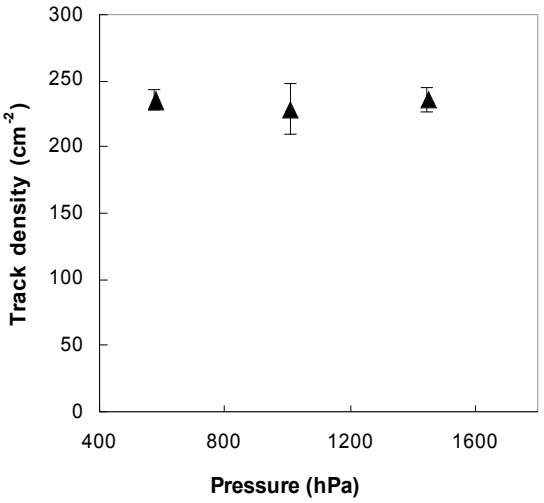

Figure 2. Effect of the pressure on the CD response.

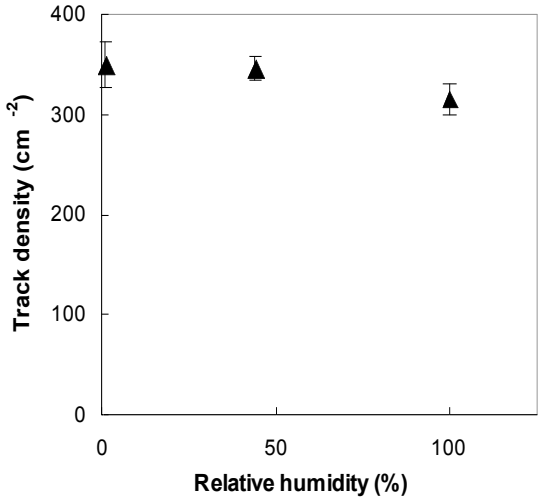

Figure 3. Effect of the relative humidity on the CD response.

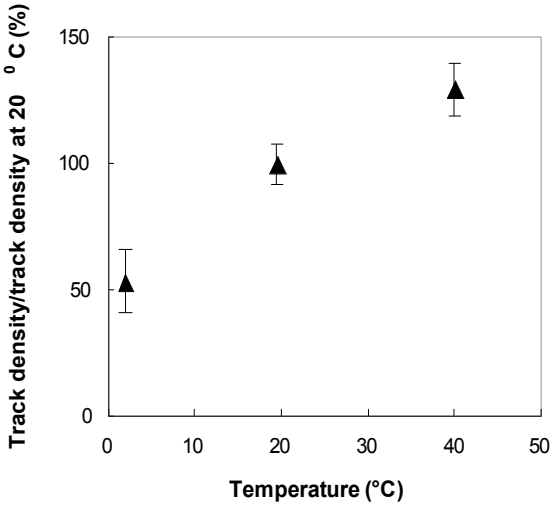

Figure 4. The effect of the temperature for CDs at depth 83 μm. The response is normalized to temperature of 20^0 C.

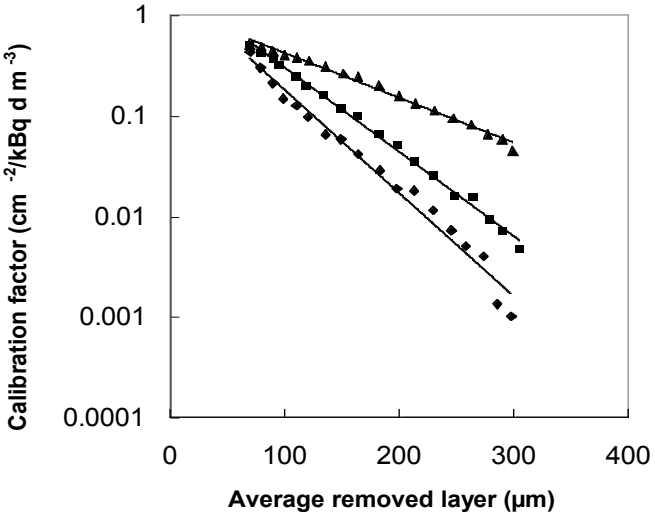

Figure 5. The dependence of the calibration factor on the depth beneath the front surface of the CD. The dependences are at (♦) 5^0 C, (■) 19.5^0 C and (▲) 38^0 C.

Comparison of CDs from different producers.

At present CDs are produced by many manufacturers. Therefore, it is important to study whether there are significant differences in their radon detection properties. Here, we present a comparison between 6 sorts of CDs from 6 different manufacturers. These were:

-Imation CD-R 650 MB 4x and 8x
-Kodak CD-R Ultima 80 700 MB
-Maxell CD-R 80XL 700 MB
-Fujifilm CD-R multispeed
-Sony CD-R 650 MB
-EMTEC/BASF CD-RW 650 MB

The first five sorts of CDs are of the types that typically can be found in the market. In the last CD the surface is covered with a very thin (< 1 μm) protective coating against solvents. As this may potentially stop or reduce radon penetration through the surface and a special design of the CPE procedure is necessary, this case will be considered separately.

All CDs were exposed to the same integrated ^{222}Rn concentration. The results are presented in Fig. 6 and do not show a considerable difference between the first 5 types. Yet our experience shows that variations in bulk-etch velocity, and perhaps in the track-etch properties, are possible for different sorts of CDs. Therefore a strong control of the removed layer and an individual calibration of different sort of CDs seems to be a good practice.

With regard to EMTEC/BASF CD-RW 650 MB, it seems that the technology involved, in which a very thin anti-solvent coating on the CD surface is used, is not common for most of the CDs/DVDs. We didn't find any other CD of such kind among the large number used in the experiments. The coating withstands the etching solution, so to remove it and to have access to the polycarbonate material an extra fine abrasive paper was used. The observed track density at a depth of 83 μm was lower than that of the other CDs by a factor of 4. The conclusion is that the protective coating stops only partly the radon penetration in the polycarbonate.

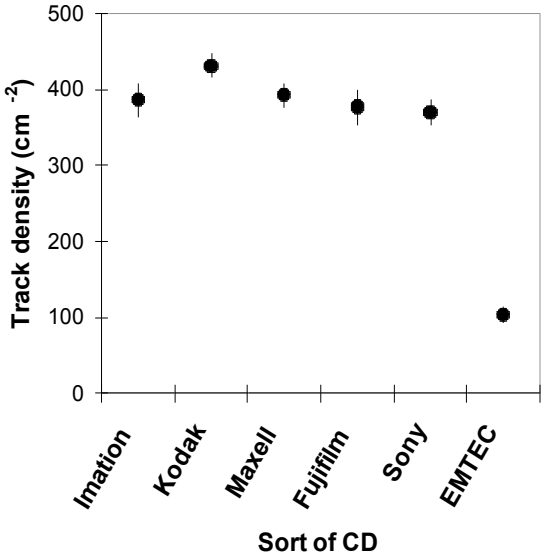

Figure 6. Comparison of different sorts of CDs.

If a CD-based radon survey program is established, an influx in the laboratory of a wide variety of CDs could be expected. The CDs should be checked for anti-solvent coating and a great difference in radon detection properties is expected only if such coating exists. Anyway, a benefit of the CD method is the possibility for an *a posteriori* calibration, in those cases where the type of CD is unknown or if there are no calibration data for them.

The useful range of the method

The calibration experiments were carried out in the range of integrated concentrations usually between 200 and 1000 kBq d m^{-3}. This range corresponds to a ten-year exposure at ^{222}Rn concentrations between 55 and 275 Bq m^{-3}. This range was selected for practical reasons, as these track-densities warrant good counting statistics and, at the same time, are well below the saturation level. However, the usable range is much wider. The lower limit of detection (LLD) was determined by using the background track density and its uncertainty according to the approach of Currie [14]. The background uncertainty σ_B was considered as a quadratic sum of two components, the first due to the counting statistics and the second due to possible differences between individual detectors. In this respect, the minimum detectable concentration for an exposure time t is given by:

$$MDC=\frac{2.71+3.29\sqrt{n_B S+(\sigma S)^2}}{CF.S.t}, \qquad (1)$$

where n_B is the background track density, σ is its standard deviation obtained after track counting of a large number of detectors, S is the examined detector surface. In respect of

CDs: the CD surface (one side) is about 100 cm^2 and there are no basic limitations to etch the entire surface, if necessary. However, in the present experiments 4 pieces per CD were etched and a 2.5 cm^2 area was counted from each, or in total 10 cm^2 per CD. The background at 83 μm was 2 ± 1 cm^{-2}.

The saturation level, using our etching and counting mode, is normally at about 2000 cm^{-2}, however, under certain circumstances, track densities up to 4000 cm^{-2} could be registered with manual counting. The useful range of the method corresponds to track densities between the LLD and the saturation level. This means that for a ten-year exposure time and ECE at 83 μm ^{222}Rn concentrations in the range 3 – 3000 Bq m^{-3} can be quantitatively measured. These estimates imply that the method covers practically the entire range of indoor radon concentrations. It should be noted that the upper limit of detection is quite flexible. At greater depths significantly higher radon levels are measurable. In this direction the reserve of CDs is large, as their full thickness is about 1.3 mm.

CONCLUSION

In the present work the performance of the recently proposed method for retrospective long-term radon measurements, by home stored CDs, is comprehended. The jewel cases, atmospheric pressure variations and dust deposition apparently have no effect on the detection properties. The effect of humidity and cigarette smoke is restricted to about 10% and this only under conditions that are unlikely to be common indoors, i.e. RH = 100% or extremely dense cigarette smoke. There are no great differences in the detection properties among the studied sorts of CDs without anti-solvent coating. The CDs with anti-solvent coating are of reduced sensitivity, but are still sufficiently sensitive to be used for retrospective measurements indoors. The factor known so far that is of major importance for the method is the temperature. Without knowledge of the temperature during the exposure time the accuracy of the method is restricted to within ± 30%, but if the temperature is known, there is a potential for precise measurements within less than 10% uncertainty.

In general, the method demonstrates a good number of benefits. It is sufficiently sensitive, not dependent on the particular radon progeny/aerosol behavior indoors, and an *a posteriori* calibration is possible. A potential for precise measurements clearly exists. Practical implementation of the method will show whether this potential could be realized.

Acknowledgement: The creative and highly reliable work of Jozef Buysse at any stage of the laboratory experiments is kindly acknowledged. The author was supported under contracts of Bulgarian Ministry of Education and Science VUF-08/05 and of National Innovation Fund IF-02-18/2005.

References

1. Harley NH, Harley JH. Potential lung-cancer risk from indoor radon exposure, Ca-A Cancer Journal for Clinicians 40:265-275; 1990.
2. Lively RS, Ney EP. Surface radioactivity resulting from the deposition of ^{222}Rn daughter products. Health Phys 52:411-415; 1987.

3. Samuelsson C. Retrospective determination of radon in houses. Nature 334:338-340; 1988.

4. Cauwels P, Poffijn A. An improved model for the reconstruction of past radon exposure. Health Phys 78: 528-532; 2000.

5. Cornelis J, Landsheere C, Van Trier A, Vanmarcke H, Poffijn A. Experiments on glass-absorbed Po-210. Appl Radiat Isot 43:127-138; 1992.
6. Oberstedt S, Vanmarcke H. Volume traps-a new retrospective radon monitor. Health Phys 70:222-226; 1996.

7. Hadley SA, Meyer NR, Fleischer RL, Cavallo A. Eyeglass lenses for personal radon dosimetry. Health Phys 79:242-250; 2000.

8. Pressyanov D, Van Deynse A, Buysse J, Poffijn A, Meesen G. Polycarbonates: a new retrospective radon monitor. Proceedings of IRPA Regional Congress on Radiation Protection in Central Europe, Budapest, 23-27 August 1999: 716-722.

9. Pressyanov D, Buysse J, Van Deynse A, Poffijn A, Meesen G. Indoor radon detected by compact discs. Nucl Instrum Methods A 457:665-666; 2001.

10. Vanmarcke H, Janssens A. Study of the properties of electrochemically etched α-tracks in a polycarbonate foil used in a radon diffusion chamber. Nucl Tracks 12:689-692; 1986.

11. Tsankov L, Pressyanov D, Mitev K, Georgiev S, Dimitrova I. Automatic counting of chemically etched tracks by means of a computer scanner. Radiat Meas 39:557-559; 2005.

12. Pressyanov D, Buysse J, Poffijn A, Meesen G, Van Deynse A. The compact disk as radon detector – a laboratory study of the method. Health Phys 84:642-651;2003.

13. Pressyanov D, Buysse J, Poffijn A, Van Deynse A, Meesen G. Integrated measurements of ^{222}Rn by absorption in Makrofol. Nucl Instrum Methods A 516:203-208; 2004.

14. Currie LA. Limits for qualitative detection and quantitative determination. Anal Chem 40:586-593; 1968.