HIGHLY SENSITIVE PASSIVE DETECTORS FOR SHORT-TERM PRE- AND POST- MITIGATION MEASUREMENTS

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Abstract

It is required to evaluate the achieved radon reduction, preferably shortly after the mitigation system is activated. As instantaneous measurements are affected by short term radon variations, few days pre- and post-mitigation integrated measurements of sufficient sensitivity is preferred. Within the European MetroRADON project novel detectors of sufficient sensitivity and with compensated temperature dependence of the response were developed. They are based on using DVDs of low intrinsic background as track detectors, covered with Makrofol N foils. The absorption of radon by Makrofol N is very high (concentration ratio foil/air is > 100). The Makrofol N foil serves as radon absorber/radiator that greatly amplifies the signal (net track-density at etched DVDs). The achievable sensitivity is sufficient to prove quantitatively (within one-week exposure) that the reduction to low radon levels (< 100 Bq m^{-3}) is achieved after mitigation. The detectors are cheap and usable for measurements at many points in large buildings. A novel technical concept (patent pending) to reduce the temperature dependence of the detectors and to eliminate the influence of thoron and humidity is described. The results of pilot experiments shown demonstrate the feasibility of this concept.

Introduction

The efficient way to reduce the radon risk in buildings with high ^{222}Rn levels is mitigation. Despite that the radon mitigation industry exists for more than 30 years, still the "mitigation outcome" can hardly be predicted and the achieved mitigation efficiency is case-specific (Kumar *et al.*, 2012; Pressyanov, 2016). The achieved efficiency and post mitigation radon levels have to be assessed in any mitigated building. The World Health Organization (WHO, 2009) recommends the reference levels, above which mitigation should be considered, to be set within the range of 100-300 Bq m^{-3}. Therefore, ideally the mitigation outcome should be radon concentrations reduced to less than 100 Bq m^{-3}. Although the ^{222}Rn concentrations before mitigation are usually above the European Union's reference level (that shall not be higher than 300 Bq m^{-3} in the Member states of the EU, according to the European Council Directive (2014)), those after the mitigation could (and should) be well below 100 Bq m^{-3}. Preferably, post mitigation levels should be evaluated by integrated measurements under conditions at which the inhabitants normally live. At the same time the mitigation contract may require verifying the achieved efficiency in a reasonably short time after the mitigation work is completed. In addition, depending on the size of the building, measurements in many points may be needed,

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which makes the use of, for example, integrating electronic monitors or electret chambers a difficult option for many mitigation contractors. In this work we studied a new design of a passive alpha-track detector in which the tracks can be etched and analyzed from a large detection area and which can provide reliable quantitative measurements at levels below 100 Bq m^{-3} within one week exposure time. The increased sensitivity is achieved by using of DVDs of low background as large area track detectors, and use of radon absorbing foils to amplify the signal, as first proposed by Tommasino *et al.* (2009). Constructively, DVDs consist of two halves stuck together, as shown in Figure (1). The front half is made of polycarbonate material that has radon absorption and track-etch properties (Pressyanov *et al.*, 2001). After mechanical splitting of DVDs, the internal surface of the polycarbonate half of the DVDs is used as the detection surface. It is covered by two foils of Makrofol N. Because of the unique radon absorption ability of Makrofol N (the radon concentration in it is 112 times higher than that in the ambient air, at room temperature (Mitev *et al*., 2016)) it serves as absorber/radiator that sufficiently amplifies the signal (net track density, *i.e.* the track density after the background is subtracted). The results from experiments and modeling that are presented below demonstrate the ability of the proposed detectors to measure low ^{222}Rn concentrations within relatively short exposure time (e.g. a week).

One limitation identified of this kind of detectors is the significant dependence of their response on the temperature. In this report we are proposing a novel concept (Pressyanov, 2019) that makes it possible to reduce or even to eliminate the temperature dependence of the detectors. The results of the pilot experiments presented below demonstrate the feasibility of this concept.

Materials and Methods

Widely preferred methods for integrated ^{222}Rn measurements employ alpha track detectors. In the commercial monitors small area track detectors are used, usually of area of few cm^2, which limits their sensitivity. Following the approach of Currie (1968) the minimum detectable average concentration (*MDAC*) after exposure time *t* is:

$$MDAC = \frac{2.71+4.65\sqrt{n_B}}{CF.t.\sqrt{S}}, \qquad (1)$$

where n_B is the background track density, *CF* is the calibration factor (*CF* = net track density /integrated ^{222}Rn concentration in the ambient air), *S* is the etched detector area from which the tracks are counted, and *t* is the exposure time. In the present study the *MDAC* is reduced by:

- increasing *CF*, by coupling the detector with an external radiator with high radon absorption ability (Makrofol N);
- increasing *S*, using DVDs as large area alpha track detectors;
- reducing n_B by using the internal surface of the DVDS, which has very low background.

In the last years the CD/DVDs method has been widely used for measurements in dwellings, caves and workplaces (Pressyanov *et al*., 2019; Dimitrov and Pressyanov; 2018, Burghele *et al*., 2017). The method employs the high radon absorption ability of the polycarbonate material of which the commercial CDs and DVDs are made and its track-etch properties (Pressyanov *et al*., 2001). The tracks created by the absorbed radon and its progeny are analyzed at certain depth (usually about 80 µm) beneath the disk front surface (Pressyanov, 2009). In the present version, which is also suited for short-term prospective measurements, the sensitivity is increased by covering the sensitive surface of the DVDs by two foils of 43 µm thick Makrofol N - a material which radon absorption ability is much higher than that of the CD/DVDs (Mitev *et al*., 2016), and etching tracks on that surface. As noted elsewhere (Dimitrova *et al*., 2011), the internal surface of the DVDs has a very low background track density: 1.1 ± 0.3 cm^{-2} (see Figure (1)) and it can be reduced to about 0.5 cm^{-2} by thermal annealing at 120 ^{0}C. In the same time the DVDs are "track detectors" of large area – up to 100 cm^{2} can be etched and the tracks from the etched area counted. The time needed the absorbed ^{222}Rn to reach 99% of its equilibrium level (with foils of thickness 43 µm) varies from several hours (at 38 ^{0}C) to about two days (at 5 ^{0}C) (Pressyanov, 2011). Therefore, the present design could be used for exposure times of one week or more and it is a good practice to dismantle and etch detectors at least two days after the end of exposure.

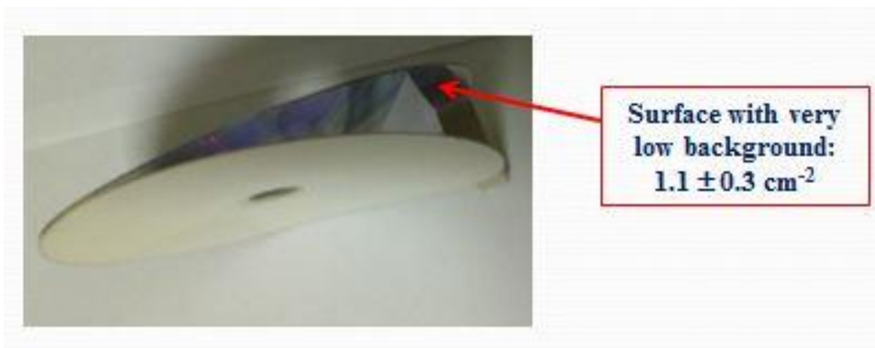

Figure (1): The structure of a DVD: it consists of two halves stuck together. The front half is made of polycarbonate which can be used as alpha-track detector. The background of the internal polycarbonate surface is very low, and can be additionally reduced by thermal annealing.

The design of the described ^{222}Rn detector is shown in Figure (2). Two DVD polycarbonate halves are stuck together with 2 foils of Makrofol N in between. Each of the detection surfaces face the absorber/radiator. The disks and foils are not stuck hermetically and radon can diffuse freely between them (Tommasino *et al*., 2009). Modeling (Pressyanov, 2009, Pressyanov *et al*., 2018) suggests that more than 80% of the signal is due to the absorbed radon in the foils of Makrofol N and the rest is due to the absorbed radon in the polycarbonate material of the disk. After exposure, the disk surface is etched electrochemically (ECE). The ECE process is performed at effective electric field of 3 kV/mm. The etching solution is a mixture of ethanol with 6M KOH solution with 1:4 volume ratio. The process starts with 30 min pre-etching with the same solution. After pre-etching the electric field is applied for 3 hours. With this ECE regime tracks are enlarged to a diameter of about 100 µm (visible by naked eye) and are usually counted by a computer scanner (Mitev *et al.*, 2010).

Figure (2): (a) Scheme of the detector element; (b) Detector element ready to use.

Results and Discussion

The proposed detectors were experimentally studied and calibrated using the dedicated exposure facility at the Laboratory of Dosimetry and Radiation Protection, Sofia University "St. Kliment Ohridski" (Pressyanov *et al.*, 2017). With these detectors we had participated successfully in the international 2017 radon intercomparison organized by Public Health England. Assessment of the *MDAC* showed that after one-week exposure the *MDAC* is less than 20 Bq m^{-3} when the entire 200 cm^2 surface of the detector element is etched and the tracks counted. If the background is reduced by thermal annealing the *MDAC* of about 12 Bq m^{-3} can be achieved after one-week exposure time. However, a problem with strong dependence of detector's response on the temperature has been identified.

The type of detectors, described above, employ radon solubility in plastics – a process which is known to depend on the temperature. Therefore we have studied the influence on the detector response of the temperature during exposure. Experiments at three different temperature levels were carried-out. The results revealed that the response of these detectors is highly dependent on the temperature. As seen in Figure (3a) the *CF* drops about 2.6 times when the temperature raises from 5 ^{0}C to 35 ^{0}C. This seems to be a significant obstacle to perform precise ^{222}Rn measurements with these detectors, when the temperature during exposure is not known and/or it may vary. The last is frequently experienced in practice when measurements are performed in different seasons in buildings which are not or are partly heated/air-conditioned.

However, a novel technical concept was proposed (Pressyanov, 2019) with a potential to overcome "the temperature dependence problem" of these, and possibly of many other types of detectors. Consider an alpha-track detector placed in a cup/chamber ("diffusion chambers") in which radon gas diffuses from outside. To protect the detector from humidity and thoron influence, many such chambers are covered by, or packed with, a polymer foil (Ward *et al.*, 1977). The foil stops radon and thoron progeny, as well as the short-lived thoron (^{220}Rn) and prevents moisture penetration. However, ^{222}Rn diffuses through the foil and reaches concentration inside the chamber that is proportional to that outside. As shown elsewhere (Ward *et al.*, 1977, Fleischer 1992), the ratio of the ^{222}Rn concentration inside the chamber (C_{in}) to that in the ambient air (C_{out}) is given by the expression:

$$\frac{C_{in}}{C_{out}} = \frac{1}{1+\lambda\frac{hV}{PS}}, \qquad (2)$$

where h is the thickness of the polymer foil, V is the volume of the chamber, S is the area, covered with the polymer foil, λ is ^{222}Rn decay constant and P is the “radon permeability” (Ward *et al*., 1977) of the material of which the polymer foil is made.

Although polymer foils are effective barriers against humidity and thoron, it has been noted that due to the temperature dependence of the radon permeability, the ratio C_{in}/C_{out} and therefore the response of these chambers to radon depends on the temperature (Fleischer *et al*., 2000; Tommasino, 2016). Figure (3b) displays that this dependence seems reciprocal to that of the detectors described in the presented report. This led to a novel, patent pending technical concept (Pressyanov 2019): designing a “compensated module” in which the detector is placed, that facilitates reduction or elimination of the temperature dependence of the detector (Figure (3c)) by selecting the parameters *h*, *V*, *S* of the module and the foil material. Pilot modeling showed that this goal is achievable, but detailed data on the radon permeability of polymer foils at different temperatures may be needed.

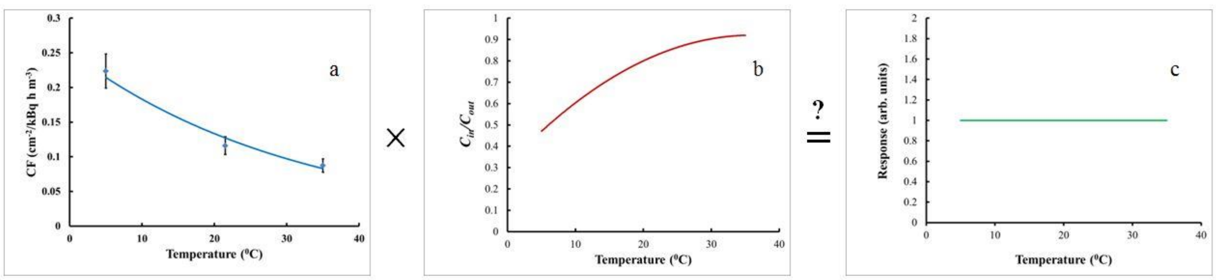

Figure (3): (a) Temperature dependence of the *CF* of detectors; (b) Typical dependence on the temperature of the ratio C_{in}/C_{out} in a volume in which radon penetrates by diffusion through plastic wall(s) (Tommasino, 2016); (c) The concept: would it be possible by placing the detectors (a) in a volume (b) to achieve compensated temperature dependence of the response.

At this stage we made a proof-of-concept study based on rather scarce data available for the permeability of low density polyethylene at different temperatures. Using the experimental results for C_{in}/C_{out} at three different temperature levels for chambers covered by low density polyethylene and interpolation between the experimental points (Pressyanov, 2019) it was crudely estimated that if the detectors described here are hermetically packed with 75 µm thick low density polyethylene, so that the ratio of the internal pack volume to the polyethylene surface is 3-4 cm, the temperature dependence would be significantly reduced. A photo of the packed detector element is shown in Figure (4).

Figure (4): Detector packed in the "compensated module".

Experiments were made at temperatures of 5 ^{0}C, 21.5 ^{0}C and 35 ^{0}C with packed and non-packed detectors. During exposure the ^{222}Rn concentrations were followed by a reference radon monitor AlphaGUARD PQ 2000 Pro (Saphymo/Bertin instruments). The results for the *CFs* of packed and non-packed detectors are shown in Figure (5). As seen the temperature dependence is significantly reduced when the detectors are packed in such package.

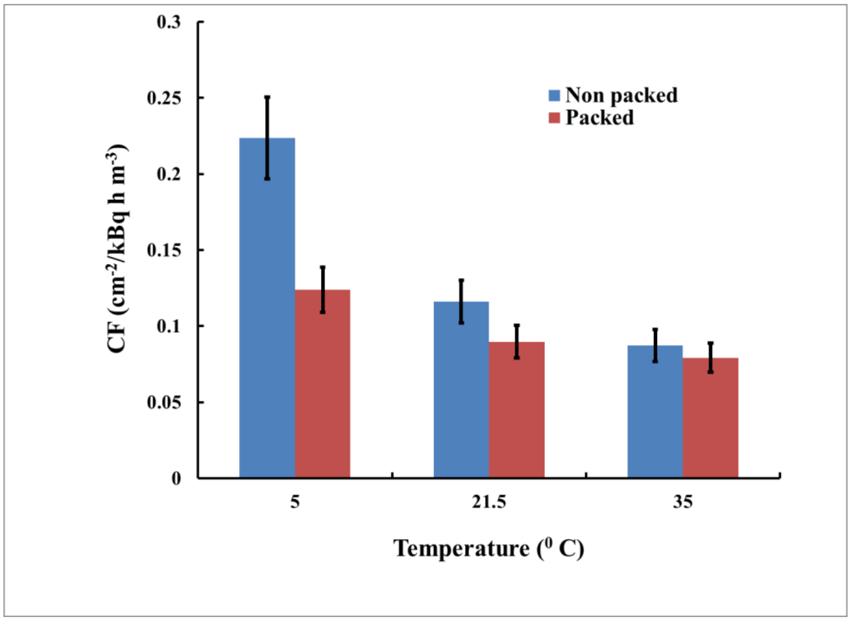

Figure (5): Dependence of the CF on the temperature of packed and not-packed detectors. The temperature dependence of packed detectors is significantly reduced.

In general, such "compensated modules" can be used with many kinds of radon detectors which response decreases with increasing the temperatures. The list of such detectors include those using activated charcoal (Cooper *et al.*, 2011), track detectors, e.g. CR-39 which show fading (fading is greater at higher temperature (Caresana *et al.*, 2010)) etc. At present extensive research work is ongoing both in the direction to determine precisely the permeability of various polymer foils over wide range of temperatures and to design compensated modules suitable for different kinds of radon detectors.

As the ratio C_{in}/C_{out} -is always less than 1, the *CF* and therefore the *MDAC* of packed detectors will be somewhat worsen. The most conservative estimate for the reduced sensitivity was for packed detectors that were exposed at 35 ^{0}C where the lowest *CF* was obtained (see Figure (5)). The results are illustrated in Figure (6), which is based on application of eqn. (1) using the experimental values obtained for n_B and *CF*. As seen, the *MDAC* is still well below 100 Bq m^{-3} if more than 20 cm^2 of the detector's surface is analyzed. When the entire surface of the detector element is etched and the tracks counted, for one-week exposure the minimum detectable ^{222}Rn concentration can be even less than 20 Bq m^{-3} if thermal annealing is applied prior the exposure of the DVDs used. Therefore, the packed detectors still fit the required sensitivity for post-mitigation measurements. In addition, the used compensated package is also an efficient barrier against moisture/humidity and thoron interference.

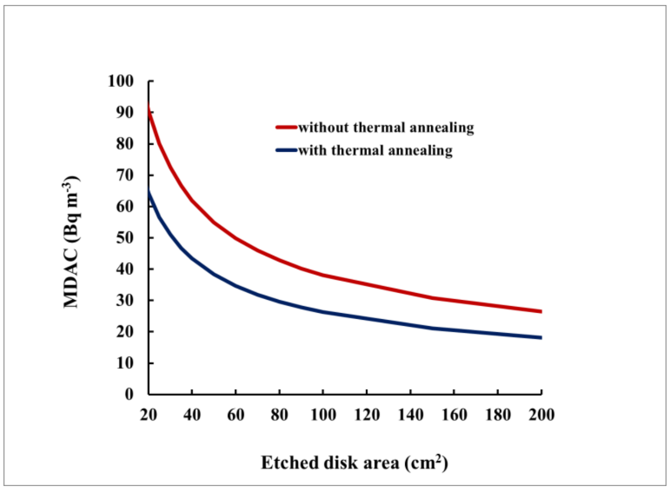

Figure (6): Minimum detectable average ^{222}Rn concentrations after one week of exposure of "packed detectors" as dependent on the surface which is etched and tracks are counted.

The detectors design described in this report provides new opportunity for application of CD/DVDs in radon industry. That is for using them for ^{222}Rn diagnostics before mitigation (Pressyanov, 2016) and for post-mitigation measurements to verify the mitigation goal is achieved. With a compensated module/package they can be used over a wide range of environmental temperatures, being protected also from humidity and thoron influence. Further research will be focused at achieving the best possible temperature compensation.

Conclusion

In this work a passive radon detector of sufficient sensitivity for post-mitigation measurements is described. It is based on DVDs used as alpha track detectors of large area, which detection surface is covered by 2 foils of 43 µm thick Makrofol N. The foil serves as radon absorber/radiator. Due to the uniquely high radon absorption ability of Makrofol N, low background and the large detection area the sensitivity of this detector is significantly increased. As a result the *MDAC* below 20 Bq m^{-3} is achievable within a one-week exposure time. With such sensitivity the detectors can be used for pre- and post-mitigation measurements. The detectors are simple, cheap (the cost of one new DVD on the market is usually less than 0.5 USD and Makrofol N foils can be used many times) and parallel measurements in many points are possible. Although the response of the detector depends on the temperature, a novel technical concept was tested. This concept allows, by placing the detector in a special package/box ("compensated module") to sufficiently reduce the temperature dependence. In addition to the temperature compensation, the "compensated module" would be an efficient barrier against humidity and thoron influence on the detector's response. Further research will be focused on improving the design of the compensated modules to achieve the best possible temperature compensation. This would open the possibility to expand the concept of temperature compensation towards many kinds of detectors, response of which depends on the temperature similarly to that of the detectors described in this report.

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