USE OF POLYCARBONATE MATERIALS OF HIGH RADON ABSORPTION ABILITY FOR MEASURING RADON

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The report comprehends one decade experience in the use of the high radon absorption ability of bisphenol-A based polycarbonates for measuring radon. The three major directions of research and development include: (1) Use of the polycarbonate material, in particular that of which CDs/DVDs are made, as both radon absorber and track detector for measuring radon; (2) Use of a polycarbonate foil as an absorber that serves as radiator of an external detector; (3) Use of CDs/DVDs for combined retrospective measurements of radon and thoron. The emphasis is made on the application of CDs/DVDs as “radon sensors” available practically at any dwelling. This allows to develop an integrated approach to the radon problem. Within this approach the basic process “detection → diagnostic → mitigation” is considered as an united whole, all actions of which can be performed in relatively short time. This is illustrated with a case of kindergarten, one of the problem dwellings identified and mitigated during our year 2012 radon campaign.

Key words: radon absorption, polycarbonate materials.

1. INTRODUCTION

Albeit radon exposure was recognized as a human carcinogen long time ago [1] the relative weight ascribed to this risk factor for population was increased in the last decade. In 2009 the World Health Organization pointed-out the radon exposure as the second cause for lung cancer, after smoking and reason number one for never smokers [2]. In 2010 the International Committee on Radiological Protection has increased almost twice the risk coefficient for radon exposure [3]. Such trends and magnitude of the radon risk need sound strategy for health risk reduction that reflects also radon measurement options.


A relatively new direction in radon measurements was introduced in the last decade. It employs the remarkable absorption ability to noble gases of bisphenol-A based polycarbonates (trade names: Makrofol, Lexan, Iupilon etc.). The materials like Makrofol were used as solid state nuclear track detectors (SSNTDs) for decades. When noted in 90-ies, their high radon absorption ability was first considered as a potential source of error in radon measurements [4]. To combine it with the track-etch properties of the material for measuring radon was first proposed in 1999 [5]. As these polycarbonates are widely used as a basic constructive material for compact disks (CDs) and Digital Verbatile Disks (DVDs) it was proposed in 2001 to use home stored CDs/DVDs as retrospective radon detectors [6]. Probably, this is the most promising application of the polycarbonate method to now, but not the only one. At present one can distinguish three main directions of development of the polycarbonate method: The first (or the “classical”) direction is that in which the polycarbonate material serves both as absorber and detector. In the second one, the polycarbonate material serves only as an absorber/radiator and the emitted radiation is registered by an external detector. The third, and the newest direction includes thoron ($^{220}$Rn). It commenced in 2011 and uses CDs/DVDs for combined retrospective measurements of the both isotopes of radon that are of radiological interest: $^{222}$Rn and $^{220}$Rn [7].

In this report the physical concepts that underlie each of the three directions are outlined, with emphasis on the use of CDs/DVDs as radon detectors. As an example for a new promising application it is demonstrated how the CD/DVD based methodology can be incorporated in an all-round integrated approach to the radon problem – from its detection and identification to the complete mitigation of dwellings of concern.

2. METHODOLOGY

2.1. CLASSICAL POLYCARBONATE METHOD FOR $^{222}$Rn

In brief, the method consists in the following: First, a polycarbonate specimen (e.g. Makrofol-foil, CD, DVD) is being exposed to radon. During exposure the $^{222}$Rn atoms that are trapped on the polycarbonate surface further diffuse in depth. After decay they emit alpha-particles and give birth to decay products, two of which ($^{218}$Po and $^{214}$Po) are also alpha emitters. As $^{222}$Rn progeny atoms rest immobilized in the polycarbonate matrix at the point of their origin, their volume distribution is the same as that of $^{222}$Rn. For a polycarbonate specimen placed in radon containing atmosphere one can distinguish “external” and “internal” alpha sources. “External” to the plastic volume are alpha-particles coming from the ambient radon ($^{222}$Rn and $^{220}$Rn) and radon progeny, as well as those from the deposited on the surface radon progeny atoms (radon progeny plate-out). The “internal” source is the absorbed $^{222}$Rn and its progeny. To avoid
influence coming from the exterior, after exposure, we remove, by chemical pre-etching (CPE), a sufficiently thick layer from the surface, and etch electrochemically (ECE) alpha tracks at that depth. If this layer is thicker than 80 µm (76 µm if take into account that α-particles of energy below certain threshold cannot create tracks) any influence of the „external“ alpha-particles is effectively cancelled out. The detailed CPE and ECE procedures and the automatic track-counting are described elsewhere [8]. The duration of the whole process (CPE + ECE + automatic counting) is about 7 hours and up to 30 polycarbonate detectors (e.g. disk specimens) can be treated in parallel.

The “signal” of the detectors is the net track density (e.g. the number of tracks per unit detector area, after the background tracks are subtracted) at the studied depth. At any fixed depth (> 76 µm) it is almost perfectly correlated with the integrated 222Rn activity concentration. Within 12 years of experimental studies it became clear that the only environmental factor known so far that can influence the signal is the temperature, but its bias can be corrected a posteriori. At present there is also a sound theoretical model that describes the process of absorption and detection in the polycarbonate matrix and that describes very well the experimental data [9].

The most promising application to date of the “classical” approach is the use of CDs/DVDs as sufficiently precise retrospective detectors [8]. Such detectors are important for the radon epidemiology as the lung cancer risk at present is formed by exposure received decades in the past [10]. This way, in future epidemiological studies the risk can be directly matched with the past radon exposure, evaluated by analysis of sufficiently old home stored CDs/DVDs. The analyzed disk should be dated to have its exposure time. The dating can be subjectively made by the disk owner or objectively – using disk marks and records. Both modes of dating are considered in Ref. [11]. Comparison between CDs and results obtained by past measurements by conventional detectors demonstrated very good correspondence [12].

Along with the epidemiological prospects, in the last 2-3 years we put the CD/DVD method in one more perspective, that could be even more important for the practice. The concept is to use CDs/DVDs as available serendipitous indoor radon sensors that can be used to pin-point dwellings with problem in short-time, possibly even on the next day after the decision to provide disk for analysis. Any disk purchased more than one year ago is suitable for that purpose. Indeed, as shown in previous studies [13, 14] the minimum detectable 222Rn activity concentration for one year exposure is about 30 Bq m⁻³. This means that with a disk that is more than one year old there is no danger of omitting buildings with radon levels increased in the range of concern (> 100 Bq m⁻³, according to the WHO criteria [2]). The probability of “false alarm” is low: The study of the background in 19 different new disks showed the average value of 6.3 cm⁻², with 18 out of 19 disks showing less than 11 cm⁻² individual track density and only one “outlier” with track density of 49.6 cm⁻² [15]. If we consider this as a representative background distribution between the new disks, there will be no “false alarm” in case of 5 years old disks and only one out of 19 with one year old disks. Such probability for false alarm (about 5% with one year old disks and lower with older
disks) is sufficiently low, moreover additional radon measurements can verify the first warning signal.

The opportunity to detect the problem even on the next day after decision to test and still this to be done by long-term integrated measurement, as recommended by WHO [2] is attractive and was tested in practice in 2012. This opportunity is discussed further in section 3.

2.2. USE OF EXTERNAL ABSORBERS AS RADIATORS

One promising direction of applications uses polycarbonate specimens only as radon absorbers and the emission from the absorbed \(^{222}\text{Rn}\) and its progeny is measured by external detector. The first attempts, in which beta/gamma detectors were used aimed measuring \(^{222}\text{Rn}\) in water [16]. Further Tommasino et al. [17, 18] expanded this approach using alpha-track detectors coupled with an absorber/radiator. Further, theoretical model of this version of the method was developed [19]. The sensitivity of such detectors appears to be between that of the CD/DVD method and the diffusion chambers with SSNTDs, traditionally employed for integrated radon measurements. However, these “radon-film-badges” as named by Tommasino [18] can be compact and easy to carry with badges, document cases etc. as personal radon dosemeters. The research and development of this version of the polycarbonate method is at the beginning step, but promising applications are visible for prospective measurements of \(^{222}\text{Rn}\) in air, water and soil-gas. The photo in Fig. 1 shows the three basic “integrated radon detectors” used in the author’s laboratory at the University of Sofia “St. Kliment Ohridski”: CDs/DVDs, radon-film badges (incorporated with the standard personal film-dosemeter badge) and the traditional diffusion chambers with SSNTD inside.

![Fig. 1 – Detectors for integrated \(^{222}\text{Rn}\) measurements used in the author’s laboratory at the University of Sofia “St. Kliment Ohridski”: CDs/DVDs, diffusion chambers (the metal can) and radon-film badges, in this case incorporated in a standard personal film-cassette dosemeter to have a personal “radon + photon + beta” dosemeter.](image-url)
2.3. COMBINED RETROSPECTIVE MEASUREMENTS OF $^{220}\text{Rn}$ AND $^{222}\text{Rn}$

In the last years the scientific community became increasingly interested in the thoron ($^{220}\text{Rn}$). Although, in some cases, the thoron exposure is non-negligible, the primary concern is the influence of the thoron presence on radon measurements [20]. In addition, until recently no methods for retrospective $^{220}\text{Rn}$ measurements have been developed. This gap was recognized by the thoron research community and the use of CDs for that purpose was suggested [21].

One possibility to expand the CD method for retrospective thoron measurements was proposed by the author [7]. The key idea is to study the signal at two depths beneath the surface. The signal in the first (which should be in the interval 64-76 $\mu$m) is created by sources, related to $^{220}\text{Rn}$ and to the absorbed $^{222}\text{Rn}$ ($R+T$ signal). The signal in the second depth (that should be >76 $\mu$m) is due only to the absorbed $^{222}\text{Rn}$ and its progeny ($R$-signal). For the purpose of measuring $^{220}\text{Rn}$ the $R$-signal is used also to determine and subtract the $^{222}\text{Rn}$ contribution to the $R+T$ signal in the first depth. The remaining “thoron component” ($T$-signal) is used to measure $^{220}\text{Rn}$. Figure 2 illustrates the concept. The sensitivity of this approach is analyzed by theoretical modeling. The results demonstrate promising potential for combined retrospective measurements of thoron and radon. This direction of studies is brand new and the future will reveal its potential for practical application.

Fig. 2 – The depth distribution of the signal in a CD. Depths ≤ 64 $\mu$m (the marked area) are not used for analysis. The resultant signal at 64-76 $\mu$m ($R+T$) is a sum of two components: $R$-related to the absorbed $^{222}\text{Rn}$ and $T$ – related to thoron ($^{220}\text{Rn}$). The signal at > 76 $\mu$m is related only to the absorbed $^{222}\text{Rn}$ and is used to calculate and subtract the “radon component” from the summary $R + T$ signal at depths 64–76 $\mu$m, as described in Ref. [7].
3. EXAMPLE OF APPLICATION: AN INTEGRATED APPROACH TO THE RADON PROBLEM

Albeit the primary goal in development of the CD/DVD method was for retrospective measurements (needed e.g. for epidemiology) the practical performance of the method suggested, that CDs/DVDs can be used to “pinpoint” radon problem. Our experience indicates, that dwellings with problem (e.g. in which the WHO level of 100 Bq m\(^{-3}\) is exceeded) would be detected by any more than one year old disk. In the last years this possibility was used to build an integrated approach to the radon problem.

The common practice to date needs long-term (> 3 months, as recommended by WHO [2]) measurements to be planned and carried-out. The results are usually available months or a year after the detectors are distributed in the studied dwellings. This “too many time” between the decision to test and eventual problem identification affects the stakeholders’ attitude and can reduce the concern about the reality of radon hazard and the need of measures to reduce it. However, a step ahead is possible, using CDs/DVDs as available, practically in any dwelling, “radon sensors”. This way, one can check almost immediately the radon situation in his house, as the disk processing takes several hours and the results can be available on the same or the next day after the disk is provided. Still, the results are based on long-term integrated measurements as WHO recommends, but concentrations are evaluated “in retrospect”.

Consider the basic process: “detection → diagnostic measurements → mitigation”. The possibility to detect the problem in short time after decision to provide one or more CDs/DVDs for analysis makes it possible the entire process to be considered as an united whole, with continuous sequence between the individual steps, and which can be controlled and supervised by single radon expert/team. Somewhat this is an analogy with the process “symptoms → diagnostic → therapy” in medicine that in most cases is supervised by one medical doctor/team, with no significant breaks between the steps.

Once the problem is detected and if the stakeholder is concerned, the next step is diagnostic measurements. This step takes normally about one-two weeks. It aims to check once more whether the alarm is real, to analyze radon distribution in the building, to find and explore radon sources and routes of entry.

This approach was tested for testing, diagnostic and mitigation of public buildings (kindergartens and schools) in the region of Kremikovtsi (one of the suburb areas of Sofia), Bulgaria. Comprehensive diagnostic measurements were organized and carried-out when a mean \(^{222}\)Rn concentration > 300 Bq m\(^{-3}\) was detected. Upon the results, the mitigation plan was prepared and mitigation steps initiated. In diagnostic step the \(^{222}\)Rn measurements were made by calibrated radonometers (RAD 7 or AlphaGUARD) and that of \(^{222}\)Rn progeny by an aerosol
Radiometer RV-4. Radon in soil gas measurements were made by diffusion chambers exposed (at 60 cm depth) for few days and radioactivity of soil/building materials was analyzed by gamma spectrometry. A description and methodological details of most of the methods employed in diagnostic is given in Ref. [22].

The third step of the process is mitigation. The choice of mitigation strategy and mitigation plan are much dependent on diagnostic measurements. The mitigation is executed by a company specialized in construction/repair of buildings under the technical supervision of a “radon team/expert” and following available technical standards [23]. Within this integrated approach the interval between the very first indication for the problem to the complete mitigation of the building can be within one-two months.

A case that illustrates this approach is taken from our practice in 2012. A disk from a kindergarten near Sofia showed $^{222}$Rn of about 1100 Bq m$^{-3}$. Diagnostic measurements confirmed the problem. Under “closed for 24 hours” conditions the measured $^{222}$Rn concentrations at the ground floor ranged in different rooms between 1200 and 2200 Bq m$^{-3}$. A decision to mitigate was taken by the local municipality and the works started immediately, under the supervision of the author. An active mitigation system with sub-slab depressurization at two points was planned. The position of the aspiration points was chosen taking into account radon distribution indoors and in soil-gas. After the system was started the concentrations dropped in all rooms to <100 Bq m$^{-3}$ (Fig. 3). The time scale of the entire process was as follows: The CD was analyzed at the end of March 2012 and the mitigation system was complete in the first week of May 2012. The total duration of the process was about 6 weeks. With the traditional approach based on prospective integrated measurements only detection step would need more than 3 months and the next steps, if any, could be even more delayed.

![Fig. 3 – Radon-222 concentration in a kindergarten before and after mitigation. The concentration averaged over the rooms on the ground floor and that at the room of maximum radon level are shown.](image-url)
4. CONCLUSIONS

In this report an overview of the potential applications of radon detection methodology based on the use of polycarbonate materials of high radon absorption ability is presented. Three major directions of research and development are outlined. Emphasis is made on the application of CDs/DVDs as detectors used to pin-point the radon problem in a dwelling. This allows to develop and test in practice a new integrated approach to the radon problem. Under this approach the entire process “detection → diagnostic → mitigation” can be considered as an united whole that can be closely controlled and the risk successfully reduced in sufficiently short time.

REFERENCES