NUKLEONIKA 2010;55(4):477–482

ORIGINAL PAPER

Radon research and practice in Bulgaria – from retrospective measurements to mitigation

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Abstract. An overview of ongoing directions of radon studies in the Faculty of Physics, St. Kliment Ohridski University of Sofia is presented. The focus is on: 1) Study and implementation of the polycarbonate method for measuring ^{222}Rn. In this respect the results from laboratory and field experience with this method are summarized. Its potential for precise retrospective measurements by home stored CDs/DVDs is emphasized. 2) Surveys in radon risk areas in the country. The approaches and results in this direction are illustrated on the example of the town of Rakovski. In this town lung cancer risk is twice increased for both sexes. Significantly high ^{222}Rn concentrations were observed in most of the houses and this can be the major factor contributing to the risk. 3) Mitigation of dwellings with high radon content. Mitigation works were recently initiated and our experience with passive radon barriers and active sub-slab depressurization systems is shared. Summarizing the results in all the three directions we conclude that there is a basis to enhance radon research and practice in the country. To be more efficient, these activities need collaboration with medical authorities, civil engineers and, especially in research, with international teams working in the field.

Key words: radon • polycarbonates • CDs • radon risk • mitigation

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Received: 17 June 2009
Accepted: 30 December 2009

Introduction

In Bulgaria there is a century-long tradition of radon measurements [8]. Since 1907 with hardly any interruption radon and later also radon progeny have been measured in mineral spas, in underground mines, in the human environment and in laboratory research. Experience from all these fields was gathered in the Faculty of Physics at St. Kliment Ohridski University of Sofia.

Nowadays, exposure to ^{222}Rn is considered the second most important cause for lung cancer among the population, after smoking [23]. Given its importance as a global health problem in the last decade a number of epidemiological studies of the population have been carried out [3, 5]. It was found that there is a statistically significant dose-response relationship between the risk and radon exposure even at average radon concentrations less than 200 Bq·m^{-3} [3]. In many countries the recommended action levels of radon concentrations, above which mitigation should be considered, are set close or equal to this value. There is a certain need for retrospective measurements, which are actually representative for the risk [4].

In Bulgaria, problems related to radon are among the main fields of research in the Laboratory of Dosimetry and Radiation Protection at Faculty of Physics, St. Kliment Ohridski University of Sofia. In this report our recent experience in the following ongoing directions is outlined:

- Measurements of ^{222}Rn by absorption in polycarbonates. Most efforts are focused on use of this method for making precise retrospective measurements by home stored CDs/DVDs, but experiments for measuring ^{222}Rn in water and soil-gas were also initiated.
- Surveys of radon risk areas in Bulgaria.
- Mitigation of dwellings with high radon content.

Measuring ^{222}Rn by absorption in polycarbonates

The method for ^{222}Rn measurement by absorption in polycarbonates was first proposed in 1999 [19]. The key concept of the method is to combine the remarkable radon absorption ability of polycarbonates like Makrofol® [7] with their track-etch properties [22] for quantitative measurements of ^{222}Rn. As the constructive material of commercial CDs and DVDs are polycarbonates equivalent to Makrofol, the method was adapted for retrospective measurements by home stored CDs/DVDs [13]. Further, the polycarbonate method has been extensively studied in a series of research [10–12]. Now, we are at the step of practical application of this method [16].

Methodology

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 them (^{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 and radon progeny, as well as those from the surface plate-out of the radon progeny atoms. The "internal" source is the absorbed ^{222}Rn and its progeny. With the method, after exposure, we remove a sufficiently thick layer from the surface so none of the alpha-particles from the external sources can reach the etched detection spot. This is done because the "external" sources are highly variable and dependent on particular behavior of radon and radon progeny in the ambient air and close to the surface. If the layer is thicker than 80 μm, any influence of the "external" alpha-particles is effectively cancelled out [10–13].

The methodology consist of the following steps:

- Exposure of specimens. The exposure times with this method in air are relatively long, e.g. a year or more. In retrospective mode the detectors (CDs/DVDs) are already exposed for a long time (years). Laboratory exposures, as well as those in water and soil gas are usually shorter (days, weeks).
- Outgassing. After exposure, the detectors are left in radon-free air to allow the absorbed radon to decay and degass for some time (about two weeks). This step is of significance only for relatively short exposure times.
- Removal of surface layer. The necessary thickness (usually about 80 μm) is removed by chemical pre-etching (CPE). This is done by an aqueous solution of 52% KOH (m/v) and 40% methanol (m/v) at 30°C.
- Electrochemical etching (ECE) is performed in effective electric field in the range (for different experiments) 3.0–3.6 kV·mm^{-1} at a frequency of 6 kHz (2 kHz in the early experiments) and temperature 25°C. The etching solution is a mixture of ethanol with 6 N KOH solution with a 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 h.
- Track counting. We employ automatic counting by a computer scanner. More details are given in Ref. [6].

One feature of this method is that two possible modes of calibration are feasible: standard and *a posteriori*. In a standard mode a set of new detectors is exposed to controlled ^{222}Rn concentrations. A specific benefit of the method for retrospective measurements by CDs/DVDs is the possibility for individual *a posteriori* calibration. For that purpose, the analyzed disk is cut into two or more pieces, one of which is exposed to controlled, "reference" ^{222}Rn concentrations. After that, the individual calibration factor is determined by the increment of track density in the piece used for calibration exposure.

Laboratory and field measurements

In all laboratory studies up to now we obtain almost a perfect correlation between the signal (net track-density) and the integrated ^{222}Rn concentration (Fig. 1). To reveal the full potential for precise measurements, substantial efforts were devoted to study the influence of different environmental factors. The results are summarized in Table 1. Virtually, none of the environmental factors

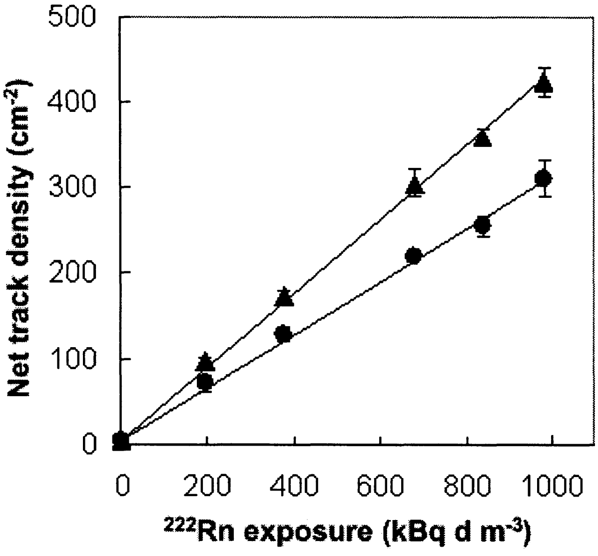

Fig. 1. Correlation between the net-track density and integrated ^{222}Rn concentration (^{222}Rn exposure). Detectors are 300 μm Makrofol foils. Track density at 83 μm (▲) and 117 μm (●) beneath the surface.

Table 1. Influence of environmental factors and storage on the performance of the polycarbonate method for measuring ^{222}Rn

Factor	Possible bias of the signal	Remark
Pressure	< 5%	Not significant effect
Humidity	< 7%	Not significant effect
Temperature	< 30%	The temperature bias can be corrected *a posteriori*
Cigarette smoke	< 5%	Not significant effect
Dust deposition	< 5%	Not significant effect
Plate out	<< 1%	At depths > 80 μm beneath the surface
CD/DVD storage in jewel cases	< 5%	Not significant effect
^{220}Rn availability	< 1%	At depths > 80 μm beneath the surface

but the temperature has a significant influence. The unknown temperature during exposure can introduce a bias up to 30%, but this can be corrected *a posteriori*. The key concept for this correction is to use the depth distribution of track-density. These distributions for temperatures 5 and 38°C (we consider the realistic indoor temperature to be in this interval) are shown in Fig. 2. As seen, the profile of the exponential depth distribution depends on the temperature. By obtaining this profile, using the signal at two or more different depths, one can reconstruct the mean temperature during exposure and this way to reduce the temperature bias to less than 10%.

The first step in the field experiments was to compare, under real conditions, the polycarbonate method with other conventional methods. Results from one of these comparisons are shown in Figs. 3a and 3b. The polycarbonate detectors were 300 μm thick Makrofol foils, while the "conventional" detectors were diffusion chambers traceable to international primary ^{222}Rn standard [9]. Very good correlation was observed for both smoking and non-smoking rooms. A good correspondence with independent "conventional" measurements was found in other comparisons, too [16].

Besides of measurements in air, in the last few years the polycarbonate method has been successfully used also for quantitative measurements in water and soil-gas [15, 16]. One benefit of this method for measurements in water is the possibility to expose polycarbonate specimens directly in the source. The potential of this approach was confirmed by the successful participation in an international ^{222}Rn in water intercomparison organized in 2009 in the USA.

Risk areas in Bulgaria

Due to the limited resources available for radon studies in Bulgaria, large scale national survey and radon mapping of the country are not yet conducted. Instead, in the last two decades the efforts were focused on relatively

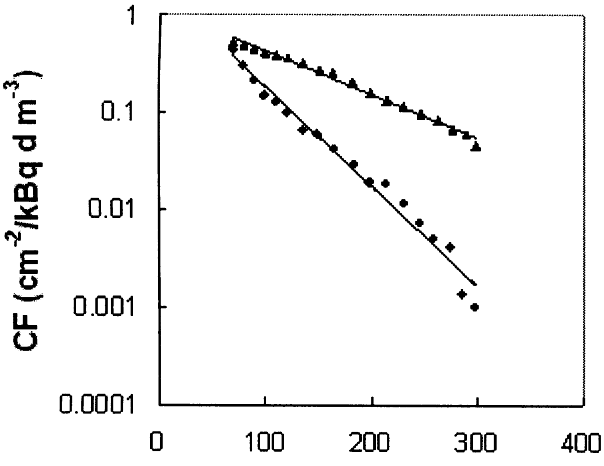

Fig. 2. Depth distribution of the response of CDs, represented by a calibration factor (CF = net-track density/integrated ^{222}Rn concentration). Results at a temperature of 38°C (▲) and 5°C (♦).

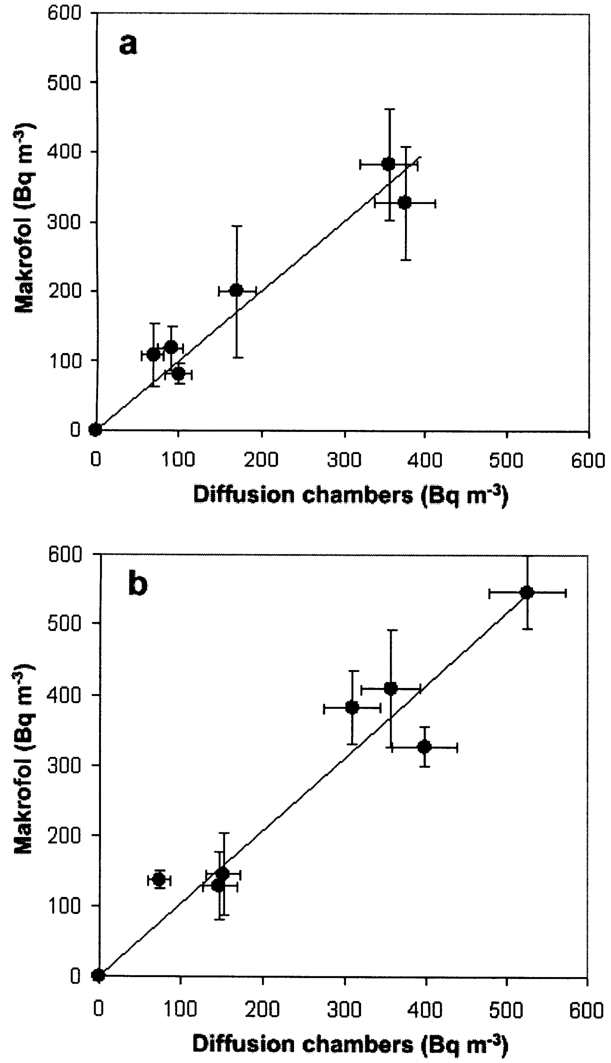

Fig. 3. Paralel measurements by polycarbonates (300 μm Makrofol foils) and diffusion chambers with Kodak-Pathe LR-115/II detectors. The exposure time was 301 days. Results are for (a) smoking rooms, and (b) non-smoking rooms.

Table 2. Cases of lung cancer in the town of Rakovski in 1981–1999 using the rates of the urban population of Bulgaria as a standard

Group	Observed	Expected	Relative risk (95% CI)*	*P*-value
Males, ≤ 49 years	34	10.28	3.31 (2.29–4.62)	$< 10^{-4}$
Males, > 49 years	140	74.36	1.88 (1.58–2.22)	$< 10^{-6}$
Males, all ages	174	84.64	2.06 (1.76–2.39)	$< 10^{-6}$
Females, all ages	27	14.19	1.90 (1.25–2.77)	$< 10^{-2}$

* 95% confidence interval.

small scale studies in "radon risk areas". Provisionaly, for working purposes, we consider a given area (e.g. town, village, region) to be risk area if the annual mean indoor ^{222}Rn concentration, averaged over the studied dwellings (but not less than 20 randomly selected), is above 200 Bq·m^{-3}. Up to now, we have revealed and regularly studied three such areas:

- Areas near Sofia, affected by uranium industry. These include the town of Bouchovo (the main center of uranium mining and milling industry in Bulgaria) and several villages nearby (Yana, Gorni Bogrov etc.). Despite that the uranium industry is now ceased in Bulgaria, parts of this region are radioactively contaminated and there are signs of the use of radioactively contaminated mining wastes for the construction of some buildings.
- Village of Eleshnitsa. This was the second center of former uranium industry in Bulgaria. The radon concentrations in this village are increased as in most of the dwellings as well as outdoors [17].
- The town of Rakovski (population about 17 000). Possibly, this town is the most interesting region for radon risk studies [18]. Significantly increased lung cancer risk is observed there for both sexes (Table 2) [14] with clearly demonstrated higher risk for young adults. Preliminary study of smoking habits in randomly selected persons revealed that the proportion of smokers in men is about the same as in the men of the urban population in Bulgaria, while in women there is a sign for lower percentage of smokers than the average proportion among urban women. The major measuring campaign in this town was carried-out in 1999–2001. At that time, about 600 integrated measurements of ^{222}Rn were made. Diffusion chambers with Kodak-Pathe LR-115/II alpha track detectors were used. These chambers were calibrated with a primary ^{222}Rn standard [9]. Most of the measurements were seasonal, with an exposure time of 3–9 months, but 157 measurements in 2000–2001 covered the whole year interval. The distribution of annual mean concentrations in dwellings, based on the whole year measurements is shown in Fig. 4. Modeling the risk suggests that radon may be the major factor responsible for the increased risk. Similar risk areas are found in other countries, too [20]. We believe it is important to target such populations for joint international epidemiological study.

Mitigation

Our experience in this field started with attempts to mitigate dwellings with very high (> 1000 Bq·m^{-3}) ^{222}Rn concentrations. Both "passive" and "active" mitigation systems have been designed and installed. We have explored two passive systems: a floor coat with special polymer putty, and a floor coverage with a soil gas retarder (200 μm polyethylene foil) with a standard floor cement coat above it. The reduction factor obtained by the passive systems was about 3. As found in other studies too [2], the reduction provided by passive radon barriers is limited and in many cases not sufficient to resolve the problem with radon. Therefore, in the last three years our efforts were focused on active systems based on active subsoil depressurization. Up to now, we have designed and consulted the construction and installation of two such systems, following documented good practices [1, 21].

Dwellings suitable for mitigation by active systems were selected following the criteria:

- The average ^{222}Rn concentration is higher than 1000 Bq·m^{-3}.
- The owner agrees to mitigate and to provide support for installing the system.

While we have documented many buildings at > 1000 Bq·m^{-3}, due to the low level of public awareness and concern about radon in Bulgaria, it was difficult to obtain the owner agreement in the second point. Anyway, in two cases we found an explicit agreement and support. In the first case we mitigated a two-floor building in Bouhovo, the ex-center of former uranium industry in Bulgaria. Before mitigation, the ^{222}Rn concentrations in this building reached 2000 Bq·m^{-3} on the ground floor. In 2007 a sub-slab sump was installed below the ground floor slab. The depressurization was ensured by an industrial in-line fan. With continuously operating system the concentrations of ^{222}Rn were reduced up to 50 times.

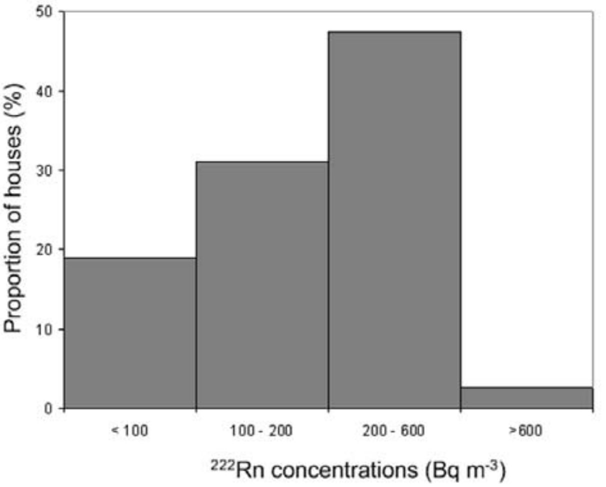

Fig. 4. Distribution of the annual mean ^{222}Rn concentrations in dwellings in the town of Rakovski, obtained in the measuring campaign 2000/2001.

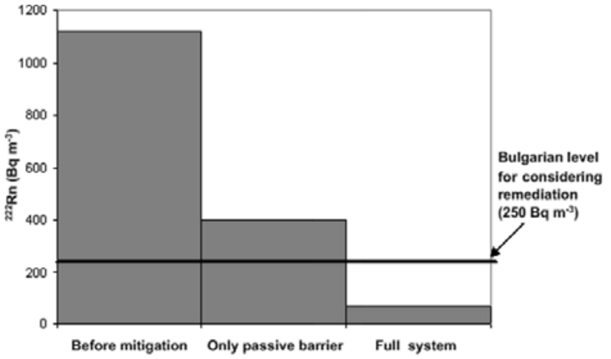

Fig. 5. Reduction of ^{222}Rn concentrations by passive barrier alone (200 μm polyethylene soil-gas retarder + 3 cm floor cement coat) and after the switching on the active sub-slab depressurization (full system).

The second case was a kindergarten in the village Gorni Bogrov (a suburb of Sofia). In this kindergarten (built in 1967) radon concentrations in the rooms ranged 800–4000 Bq·m^{-3}. The radon problem there was revealed in 2007 by occasional measurement of a CD and a diffusion chamber (as part of our research in randomly selected buildings), and consequent detailed measurements in all rooms by diffusion chambers and a radon monitor AlphaGUARD (Genitron GmbH, Germany). This problem attracted some public interest and concern in Bulgaria. As a result, the regional municipality engaged the author to provide technical consultancy for construction and installing a proper mitigation system in this building. The mitigation problem was additionally complicated by the high level of ground water beneath the building. This hampered installation of a sub-slab sump, similar to that used in the first case. The proposed technical decision was to ensure the sub-slab depressurization by two loops of perforated pipes placed in trenches. Such systems are described in the ASTM standard [1] whose rules were followed in the system design. After placing the pipes, the trenches were filled with gravel. The floor was covered by a 200 μm polyethylene foil serving as a soil-gas retarder. After that, the foil was covered by a standard cement floor coat. The depressurization of each loop was ensured by a 70W continuously working noiseless fan. Continuous night and day measurements of ^{222}Rn with AlphaGUARD were made first before the start of the mitigation work, and the second after passive barrier (polyethylene + cement coat) was placed and finally with the full system installed and running. The results, illustrating the performance of the system are shown in Fig. 5. As seen, the passive barrier alone reduces the concentrations but not sufficiently. The radon problem in this building is eliminated only after activation of the full mitigation system.

Conclusions

In this report an overview of three important directions of the present radon related work in Bulgaria is made. The first is research and implementation of the polycarbonate method for ^{222}Rn measurement, including for retrospective measurements by home stored CDs/DVDs. The second is studying the radon problem in radon risk areas. The third and the most recent one is the mitigation of dwellings with very high ^{222}Rn levels. In all of these fields results are achieved that give the basis to continue and enhance the activities. To be even more significant the progress needs closer collaboration with medical authorities, civil engineers and, especially in research, with international teams working in the field.

Acknowledgment. The first pioneering works in the polycarbonate method would be hardly possible without the enthusiasm and the highly reliable laboratory work of Jozef Buysse. In the last years K. Mitev, I. Dimitrova and S. Boychev actively contribute to the progress in this field. The work was supprted by the Research Fund of St. Kliment Ohridski University of Sofia under contract 142/2009 and by the National Research Fund.

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