AIP | Conference Proceedings

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Citation: AIP Conf. Proc. **1544**, 130 (2013); doi: 10.1063/1.4813470
View online: http://dx.doi.org/10.1063/1.4813470
View Table of Contents: http://proceedings.aip.org/dbt/dbt.jsp?KEY=APCPCS&Volume=1544&Issue=1
Published by the AIP Publishing LLC.

Measuring Radioactive Noble Gases by Absorption in Polycarbonates and Other Organics: From Radon Indoors to Nuclear Safety

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Abstract. The report summarizes recent research and practice of using materials with high absorption ability to noble gases to measure their radioactive isotopes. Most of the studies employ bisphenol-A based polycarbonates, because of their remarkably high absorption ability to noble gases. This is the material of which commercial CDs/DVDs are made and they may serve as serendipitous, already available in dwellings, radon and thoron detectors. We present the essence of the gathered experimental evidence that the CD/DVD method can successfully address some long-lasted problems in radon dosimetry: The first is making sufficiently precise retrospective ^{222}Rn dosimetry for the purposes of epidemiological studies and risk estimation. The second is rapid identification of buildings with radon problem. We demonstrate how this can be used to develop an integrated approach to the radon problem. Within this approach detection, diagnostic and mitigation are considered as an unified whole, and the interval between the decision to provide disks for analysis and the complete mitigation of the building, if radon problem is identified, is short. Besides radon and thoron, bisphenol-A based polycarbonates were successfully used to measure ^{85}Kr and ^{133}Xe for the purposes of the effluents control and nuclear safety of nuclear installations. The perspectives to employ other organic materials in which noble gases are highly soluble for measurement of their radioactive isotopes are also discussed.

INTRODUCTION

Some challenging problems in nuclear science are related to radioactive isotopes of noble gases. The list should be started with radon: In 2009 the World Health Organization (WHO) pointed-out the exposure related to ^{222}Rn as the second cause for lung cancer, after smoking, and the reason number one for never smokers [1]. In another direction – the artificial radioactive isotopes of krypton and xenon are important indicators for technological control and safety of nuclear installations [2]. The isotopes ^{85}Kr and ^{133}Xe can be usable for monitoring the Nuclear Non-Proliferation Treaty and the Comprehensive Nuclear Test-Ban Treaty [3]. Half of the radionuclide stations of the Comprehensive Nuclear-Test-Ban Traty Organization (CBTBO) shall be equipped to detect xenon. The measurement of radioxenon isotopes is of key importance after a major nuclear accident. However, one lesson from the Fukushima Dai-ichi accident was that one problem can be not in low levels measurement by sophisticated equipment and methodology, but rather in the ability to measure high levels in sufficient number of points at short distances from the place of the disaster [4].

The noble gases are soluble in many organics. The process of radon absorption in polymer materials has been studied both as a potential source of error [5] and with the goal to develop new monitors [6,7,8]. First in 1999 it was proposed to employ the remarkably high radon absorption ability of bisphenol-A based polycarbonates (trade names: Makrofol, Makrolon, Lexan etc.) for measuring radon [6]. Several years later these materials were successfully used to sample and measure ^{85}Kr and ^{133}Xe both in laboratory and real conditions [9,10]. In all of these directions the "polycarbonate method" demonstrated sufficient potential for practical applications. The most prospective is the use of

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AIP Conf. Proc. 1544, 130-137 (2013); doi: 10.1063/1.4813470

CDs/DVDs as retrospective radon detectors. These widely available disks are made of the same kind of polycarbonate material and each home stored CD/DVD can serve as sufficiently precise retrospective radon detector. The report is focused mostly on this method, but the achievements and perspectives in other directions are also discussed.

METHODOLOGY

In general, the polycarbonate-based methods are classified in two groups:

- "Classical" polycarbonate method. This version is suitable only for alpha-emitting noble gases (^{222}Rn and, more recently, ^{220}Rn too [11]). It combines the radon absorption ability of the polycarbonate material with its track-etch properties. The version for ^{222}Rn (which is already put in practical use with CDs/DVDs as radon detectors) consists in the following: After exposure, a layer from the surface (should be > 76 μm, but usually it is ≥ 80 μm) is removed. This is done to cancel-out the influence of surface defects, deposited on the surface ^{222}Rn and ^{220}Rn decay products and other "external" to the polycarbonate volume alpha sources. The experimental studies revealed that the signal (net track density) at the target depth is almost perfectly correlated with the integrated ^{222}Rn concentration [12]. At depth ≥ 76 μm the results are not dependent on thoron. Usually, the scheduled surface layer is removed by chemical pre-etching. After that the tracks at the studied depth are revealed by electrochemical etching and counted visually or automatically. These procederes are described in detail elsewhere [13, 14].
- In the second version of the polycarbonate method, the polycarbonate material serves as "absorber/radiador". It is used to sample radioactive noble gases. Further, the emitted radiation is measured by external detector. This version is usable for all radioactive noble gases, but it may require information about the kynetic of the sorption/desorption process [15]. Up to date detection by HPGe gamma-spectrometry, low-level beta counting [16], liquid scintillation counting [17] and external alpha-track detectors [8] were successfully used.

Fourteen years were devoted to study the performance of the method and the influence of different factors. The research output is given in many papers in the scentific literature (see e.g. [13] with references). The factors that were studied include:

- Pressure (within 0.5 – 1.5 atmosphere);
- Humidity (0-100%);
- Temperature (2 – 40^0 C);
- Dust deposition;
- Ciggarette smoke;
- Mode of storage of CDs/DVDs (bare, in jewel case, in envelope etc.);
- Influence of thoron (^{220}Rn) on the signal at > 76 μm;
- Effect of ^{210}Po growth in the detector volumen alter long exposure times;
- Detector fading (for up to 20 years);
- Effect of laser light used t oread/write CDs/DVDs.

In brief, all environmental factors but the temperature have no significant influence and that of temperature can be corrected *a posteriori*, as described elsewhere [12].

A theoretical model of the physical processes, envolved in the method, has been developed. Using the model, theoretical exppressions were obtained that describe:

- The process of sorption and desorption [15] (Fig. 1);
- The volume distribution of the activity – as under equilibrium (Fig. 2) as well as under dynamic stage (Fig. 3). These distribution were modelled at different ambient temperature;
- For the "classical" version of the method – the response of the material as alpha-track detector was also studied and modelled [18].

Very good agreement between the theory and the experiment was observed. Based on the theoretical model a method was developed, to detemine the partition coefficient (the term "partition coefficient" is defined as the ratio of the concentration of the gas in the material, at its surface, to the concentration in the ambient media (e.g. air, water etc.) and the difusión coefficient of the radioactive noble gase isotope in the material [18, 19]. This method is suitable for any material for which the process of sorption/desorption is ruled by classical difusión equation with radioactive decay. We have determined the solubility (evaluated by the dimensionless partition coefficient) of radon in different materials. The results (part of them obtained within international intercomparison) confirmed the remarkably higher solubility of radon in bisphenol-A polycarbonates than in other studied plastics. At room temperature we have obtained partition coefficient of Makrofol DE of about 25. Reports in the literature and our recent experimental studies suggest that it is even higher for Makrofol N (about 40) [8].

Important feature of the classical version is the possibility for individual *a posteriori* calibration. This is especially valuable for the practical application of the CD/DVD method. The concept of *a posteriori* calibration is to take a piece from the analyzed disk (CD or DVD) and to expose it additionally to reference ^{222}Rn exposure. After that both pieces (exposed and not exposed *a posteriori*) are etched simultaneously and the increment of the signal, divided by the reference ^{222}Rn integrated concentration gives the individual calibration factor (CF). In most cases the individual CFs of different brands of CDs/DVDs differ within 15%, but sometimes differences of up to 40% were found. Experimental estimates showed that the range of sensitivity of the CD/DVD method starts at 3 Bq m^{-3} for 10 years of exposure time (or 30 Bq m^{-3} for one year) and covers the entire range of indoor ^{222}Rn concentrations [20].

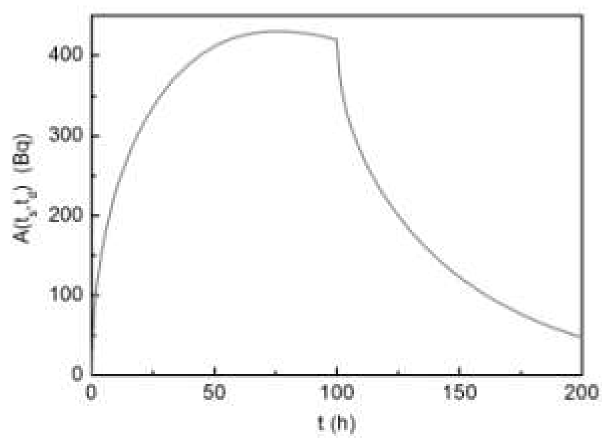

FIGURE 1. Theoretical dependence of sorption of ^{133}Xe (for 100 hours) and desorption in 200 μm Makrofol foil of 100 cm^2 area. Sorption is in hermetic volume, at initial ^{133}Xe activity concentration 10^8 Bq m^{-3}, decreasing during exposure by the radioactive decay of ^{133}Xe. Desorption is free in air. Activity absorbed in the plastic is shown [21].

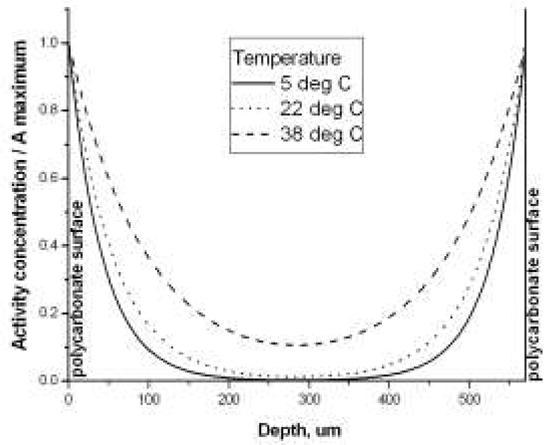

FIGURE 2. Equilibrium distribution of ^{222}Rn in a polycarbonate foil of thickness 570 μm at 3 different temperatures [22].

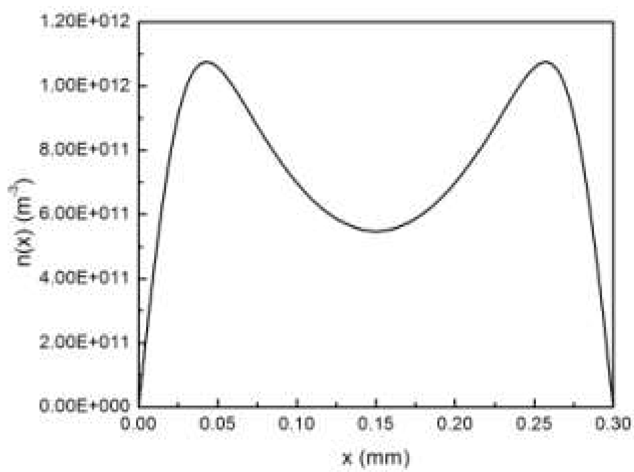

FIGURE 3. Distribution of ^{133}Xe atomic concentration in 300 μm Makrofol plastic 5 h after the end of exposure [21]. This is a transitional distribution at a dynamic stage (desorption). The preceding sorption stage was for 100 h at 10^6 Bq m^{-3} activity concentration of ^{133}Xe.

APLLICATIONS

CDs/DVDs used in retrospective dosimetry

The cancer risk due to radon is formed by exposure received for years/decades in the past. Current scientific knowledge revealed that exposure within the interval 5-35 y prior to the clinical manifestation of lung cancer can contribute to the risk [23]. Making a realistic estimate of this past exposure is in the focus of the retrospective radon dosimetry. This problem has attracted a lot of attention in the last decades and several methods were proposed. In our recent overview of the state-of-the-art in retrospective radon dosimetry [20] we point-out that the CD/DVD method can adequately address this problem for the following reasons:

- It can cover sufficiently long time in the past;
- The ^{222}Rn concentration is obtained with reasonably good uncertainty. Actually, the most important factor for the uncertainty budget – disk dating, can be successfully addressed by combining subjective dating, provided by the disk owner. with dates on the disk labels/records and by analyzing more than one disk from a house [24];
- The method is suitable for large-scale measuring campaigns [24];
- The range of sensitivity covers the whole range of indoor ^{222}Rn concentrations;
- The method is applicable in case of change of residence;
- Recent findings suggest that thoron (^{220}Rn) can be also measured in retrospect, using the signal at two depths, one in the interval 64-76 μm and another ≥ 76 μm [11].

Normally, in the practical surveys, the "disk age" was provided by its owner. A bias is possible in this "subjective" mode of dating. We explored the possibility to use record/production date of the CDs/DVDs (excluding rewritable disks) as a more objective manner of dating. Analysis of the results from the comparison of both modes suggests [24] that in 88% of cases both dates coincide within 10% and in the another 12% they differ more (by up to a factor of 3). A possible approach to reduce the probability for dating bias is to ask for 2 (or more) disks for analysis. If the disks give similar results, the probability for them to be equally biased is low (e.g less than 5% for 2 disks and even lower for more than 2 disks). If there is a significant difference, the case is resolved by additional measurements. Our experience with analysis of several disks from one place indicates, that in most cases the results are statistically similar.

CDs/DVDs used for rapid identification of buildings with high ^{222}Rn

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 warning 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 [1], and preferably one year) measurements to be carried-out. The results are usually available months or a year after the start of the measuring campaign. This "too many time" between the decision to test and eventual problem identification affects the home owners' 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, the radon situation in a house can be checked rapidly, 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 the concentrations are being evaluated "in retrospect".

Any disk purchased more than one year ago is suitable for that purpose. Indeed, the minimum detectable ^{222}Rn 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 that are in the range of concern (> 100 Bq m^{-3}, according to the WHO criteria [1]). 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} [22]. If this is considered as 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 ones) is sufficiently low, moreover additional radon measurements can verify the first warning signal.

The possibility to detect the problem in short time after decision to provide one or more CDs/DVDs for analysis makes it possible for the entire process **"detection/identification → diagnostic measurements → mitigation"** to be considered as an unified whole, with continuous sequence between the individual steps, and which can be controlled and supervised by a single radon expert/team. This makes the process similar to 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. This is an integrated approach: once the problem is identified (by CD/DVD) and if the stakeholder is concerned, the next steps are diagnostic measurements and mitigation (Fig. 4). This approach was used in testing, diagnostic and mitigation of public buildings (kindergartens and schools) in some suburbs of Sofia, Bulgaria. Once ^{222}Rn concentration > 300 Bq m^{-3} was detected comprehensive diagnostic measurements were organized and carried-out. Upon the results, the mitigation plan was prepared and mitigation steps initiated. The time needed to complete the whole process can be ½ - 2 months, according to our practical experience. The results from the year 2012 campaign in suburbs of Sofia, Bulgaria are shown in Fig. 5.

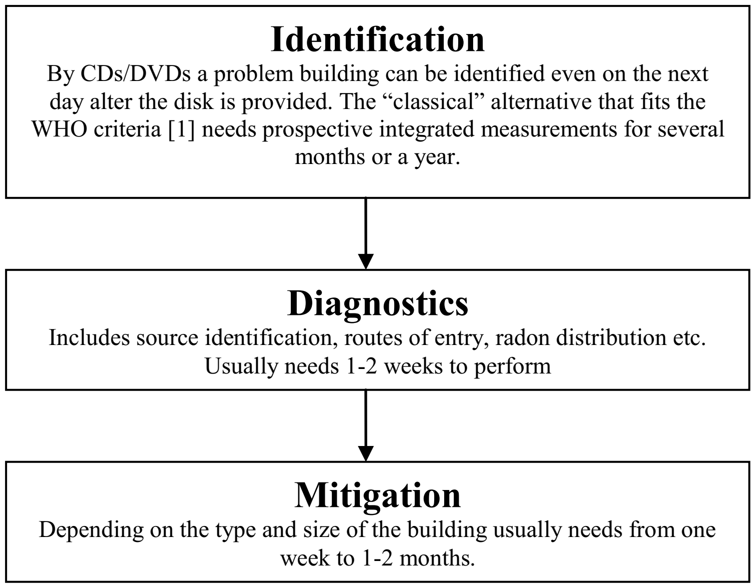

FIGURE 4. Basic steps of the integrated approach to the radon problem. For sound identification of radon problem an annual average of radon concentrations should be evaluated. With conventional methods this needs prospective exposure of detectors for long time. With CDs/DVDs this can be done in short time, even on the next day is technically possible.

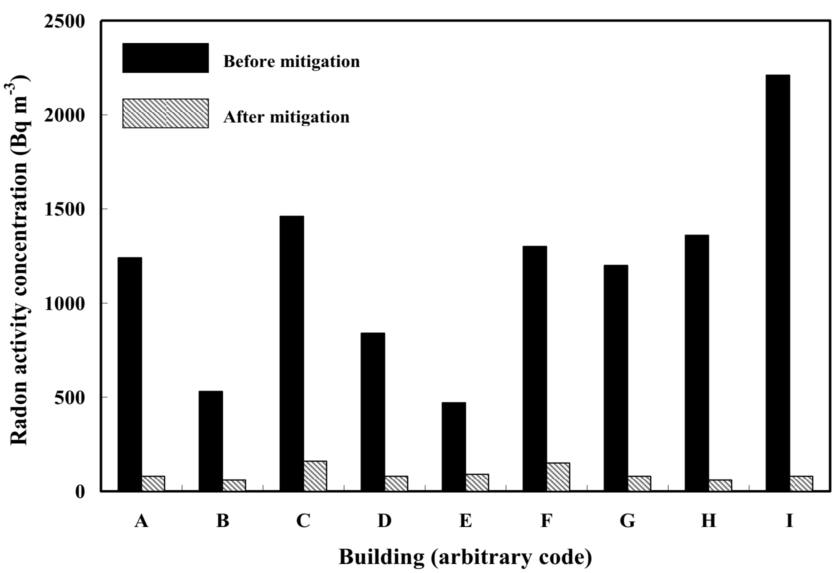

FIGURE 5. Integrated approach at work. Results of the 2012 “identification-diagnostic-mitigation” campain in suburbs of Sofia, Bulgaria. In some buldings (e.g. H and I) the whole process was completed within 1-1.5 months.

Another promising application is to detect large changes in radon concentrations in the past [24]. The concept is to analyze two or more disks of different "age". We have studied this possibility by numerical simulation and experimentally. The results suggest that changes of the order of 50% that happened in the past are detectable. This can be usable to evaluate: 1) The effect of mitigation, conducted in the past; 2) The effect of past energy-efficient reconstruction of the building on the indoor radon concentrations.

Measurement of ^{222}Rn in water

The radon absorption ability of polycarbonates was employed also to measure ^{222}Rn in water [16, 25]. In this case the measurements are prospective: The polycarbonate specimen is left for exposure that is usually short-term (days or weeks) and afterward measured. Specific benefit in this case is that the specimen can be exposed directly in the water source. The detection limit with the "classical" version of the method is about 2 Bq L^{-1} for one day exposure time. With this method we have successfully participated in an international radon-in-water laboratory intercomparison [26].

Measurement of artificial radioactive noble gases

In the last years absorption in polycarbonates was used successfully to measure ^{85}Kr and ^{133}Xe [9,10]. Note that only the second version of the polycarbonate method can be applied for artificial radioactive noble gases. The polycarbonate specimens (foils or granules) were used as samplers and the radiation from the absorbed radioactive gases is measured by external detector, usually after exposure. Measurements in air and water were made, both under laboratory and real conditions. In respect of ^{133}Xe, by using gamma spectrometry with HPGe detector of 25% relative efficiency a detection limit of 100 Bq m^{-3} can be achieved [27]. Notably, after the Fukushima accident, one problem in ^{133}Xe monitoring was at levels $\geq$ 100 Bq m^{-3}, as the only CTBTO network station that measure ^{133}Xe in Japan – that in Takasaki, reached the high dynamic range of the system at these levels and could not provide reliable results [4]. In contrast much lower levels, down to the atmospheric background, were detected in many stations at long distances, but direct information on high levels in the close vicinity of Fukushima was missing [4].

DISCUSSION AND CONCLUSIONS

One possible key phrase for the described approach is "serendipitious radiation detection", in the sense used by Fleischer [28]. This is to employ in survey of radiation and radioactivity (especially in air, where time variations can be large and rapid) suitable materials that just happen to be there and which "gathered" signal that, when analyzed, brings important information about the radiation/radioactivity levels. To date bisphenol-A based polycarbonates have demonstrated sufficient potential for practical use in measuring radioactive noble gases. This role they play because of their remarkably high, among other studied organics, absorption ability to noble gases. One method – that of the home stored CDs/DVDs used as retrospective radon detectors is already put in practical use. However, there is no reason to exclude other organics from possible applications. For instance natural and synthetic oils have been already used to measure radon [29]. We foresee a great potential to employ "serendipitous" detectors around nuclear installations. For instance after a nuclear accident with atmospheric release of large amount of radioactivity, analysis of some organics (CDs/DVDs, in particular) can bring important information about the release of xenon and krypton isotopes. At present we have focused an interest and started pilot research on whether, after an accidental radioxenon release, the radioactivity absorbed in the body of an accidentally exposed man can be used to reconstruct the levels to which the person has been exposed. The heuristic reasons are: decades long application of ^{133}Xe in nuclear medicine and in vivo measurements of nuclear workers [30] indicate that after a person is exposed to ^{133}Xe atmosphere, the body retention of xenon shows fast and slow component. The last is probably due to xenon dissolved in fat tissue. Reports (see e.g. [30]) indicate that after exposure to emergency high levels (of the order of 10^8 Bq m^{-3}) the man "holds" a measurable (by whole body counting systems) amount of ^{133}Xe in his body for several days ("slow" component). Of course, to make this concept a working approach a hard dedicated work in modeling and calibration should be done. But this just indicates that in the direction of the "serendipitous radiation measurements" we are at the beginning of the road.

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