



Radon in Workplaces the Urgent Need of New Measurements and Devices

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ABSTRACT

The existing passive radon monitors, their relative calibration facilities together with the past inter-comparison exercises have been mission-oriented towards radon measurements in dwellings. These monitors have been successfully applied throughout the world for radon measurements in homes, characterized by temperatures in the range from 20 to 25°C and a relative humidity less than 50 R.H. A multitude of different problems may arise when these passive monitors are used in environment other than homes, such as in soil and in workplaces, where large humidity up to 100 RH and temperatures anywhere from 0°C to 40°C may be encountered. Under severe environmental conditions, different measurement errors may occur which have remained concealed to date. These errors may be caused by a drastic change of both the radon diffusivity through the and for the monitor housing respectively. permeation membranes or the radon absorption by the plastics, used for the track detector. For the compliance to the assessment of the occupational exposures, it is necessary to eliminate all the possible sources of errors, which may be conducive to litigation. Another important shortcoming of the existing passive monitors is the difficult to turn them on/off daily, as required for radon measurements in workplaces. Finally, most of the problems, listed above, can be solved by the exploitation of a new generation of passive monitors, known as Rn film-badges. These monitors are similar and often identical to neutron film-badges, which have proved to be very successful throughout the world for the personnel neutron dosimetry. In particular, the present paper will describe the unique characteristics of these radon film badges, such as compactness, fast time response, any desired response sensitivity, simplicity in turning them on and off, etc.

1. Introduction

A new generation of radon monitors has been recently developed, based on radon sorption in solids [1]. Since there is already a multitude of different well-established radon monitors [2], the development of new detectors is justified only if they make it possible to carry out measurements, which are difficult, if not impossible, to carry out with existing technologies. As already proved, these newly-developed monitors present unique characteristics for the correct measurements of radon in soil and in water with concentrations from a few kBq/m³ to tens of thousands kBq/m³ [3]. In particular, these monitors, known as the radon film badges, show promise for the assessment of the radon exposures in workplaces. These occupational exposures to radon can't be simply obtained by the passive monitors, typically used for indoor measurements, especially because of their limited response sensitivity, their long response-time, and the difficulty to turn them on and off.

To this end, it is interesting to report what John Harley [4] had to say about using the same approach of the indoor radon-monitoring for the assessment of occupational exposure to radon: "If a health physicist were to recommend monitoring the exposure of workers by placing a single detector in the middle of a nuclear facility, he would be removed in a straightjacket. When we do the same thing in a house, everyone agrees. So keep in mind that, even with the best of instruments, we may not be monitoring the right thing in the right place".

Any attempt, made in the past, to use the existing passive monitors in environment other than dwellings has encountered many difficulties. For example, since 1981, it was demonstrated how poor were the characteristics of the cup-type diffusive chamber for the assessment of the occupational exposure in mines [5]. The response of this monitor was affected by the atmospheric pressure and/or altitude [6] and by the chamber design and geometry. Finally, Frank and Benton [5] proved that most

of these shortcomings could be overcome by keeping the maximum dimensions of the chamber less than 2.5 cm and by using a conductive detector holder.

In yet another important example, the passive Rn-monitors, developed for indoor measurements, proved to be drastically affected by the humidity and the presence of thoron, when used for in-soil-radon measurements. Eventually these problems have been solved by using a permeation-type of sampler [7,8]. Among all the existing passive monitors, the permeation samplers are considered to be the least affected by ambient conditions, including humidity, air current, temperature, the presence of thoron, etc. [9, 2]. However, these conclusions are true for radon monitoring in homes, but they have proved to be totally wrong for radon monitoring in workplaces, which may be characterized by a wide range of temperatures (0-40°C) and humidity up to 100% RH. [10,3]. Unfortunately, the widespread calibration facilities and the many international inter-comparisons, carried out since the 1980s [11] have been all mission-oriented toward home-environmental conditions with temperatures between 20-25°C and low relative humidity (namely 30% RH). For these reasons, inter-comparison exercises run under field conditions (humidity up to 100% RH and temperatures from 0°C to 40°C), are very valuable to identify the shortcomings of existing Rn-monitors, when used in environments other than homes, such as in workplaces, etc.

2. Concealed Errors in Radon Measurements by Permeation-Type Monitors

A permeation-type of passive radon monitor is typically formed by a track detector facing an air radiator, into which radon enters from the outside air through a non-porous membrane. The entry of radon gas into a cup, covered by a membrane [7], is characterized by a mean permeation time, τ_M , given by:

$$\tau_M = dV/PA \quad (1)$$

where V is the volume of the cup, d and A are the thickness and area of the membrane, while P its radon permeability.

Because of the large volume of the cup (350 cm³), the permeation time of the radon into the cup-type monitor is about 1.5 days. As mentioned earlier, the air radiator facing the track detector must have a thickness less than about 2,5 cm [5], resulting in air volumes of 30-50 cm³, as that of the radon monitors, developed by Tommasino et al. [8] and by Gilvin and Bartlet [12] at NRPB (now HPA) respectively. In practice, these monitors have a volume of about 40 cm³. Permeation samplers with smaller volumes are of little interest, because of them to low response

sensitivities, especially for radon measurements in dwellings. The best geometry of a permeation sampler is formed by a heat-sealed bag [8], typically formed by a polyethylene film with thickness of about 40 microns. The permeation time, τ_M , of the radon into the 40 cm³-bag of a 40 μ m-thick PE-membrane is just a few hours, as derived from equation (1). Recently, Miles et al [13] have developed a new Rn-permeation monitor, τ_M , formed by a bag with a 200 μ m-thick polyethylene film to encapsulate the NRPB monitor. Because of the large bag-thickness, this sampler has a Rn-permeation-time, τ_M , of about 1.3 days according to equation (1). Samplers with such long permeation times have been extensively applied in the past [2]. The basic principles of the permeation samplers were considered well established experimentally and theoretically [7,12,13,14]. Provided that the permeation rate into and out of the enclosed volume remains constant, the integral of the radon gas concentration within the volume, C_{in} (measured sufficiently long after the end of the exposure period) is proportional to that of the outside, C_o . In short, by keeping the radon monitor within the sealed bag after the exposure to the radon is ended, the additional decay of the trapped radon just makes up for that which was earlier missed because of the delay in entering the bag.

Unfortunately, what were the basic principles for radon monitoring are now known to be totally wrong simply because it has been discovered that the Rn-permeability of the barrier membranes (namely polyethylene) changes drastically versus the temperature. For this reason, passive radon monitors with such long radon-permeation time may be affected by different errors, which have remained concealed in the past [10,3]. These errors can be considered negligible for radon measurements in homes, where the environmental parameters are like those used in the calibration facilities. By contrast, large errors may occur when using radon monitors with a day-long permeation times for workplaces, since the response of these monitors at 0°C is a factor of three lower than that at 40°C [3]. Moreover, passive monitors with such long permeation times are all but acceptable for workplaces, where the daily occupational exposures last 8 hours (or less) a day. Finally the newly developed radon monitors, known as the Rn film-badges, make it possible to overcome most of the shortcomings of the existing radon-monitors for the assessment of the occupational Rn-exposure.

3. Film-Badges Based on Radon-Sorption Processes

In general, the uptake of any gas by solids has been termed "sorption" by McBain [15] to include absorption and adsorption as special cases. The Rn-sorption processes have been extensively exploited in the past for the sampling of

radon gas and for its extraction from other gases and/or liquids [16]. In particular, the liquid scintillators, because of their radon absorption characteristics have been widely used for decades [17] for water-borne radon measurements. The radon absorption in both liquids and non-porous (dense) polymers is based on the so-called “solution-diffusion theory”, according to which, the absorption processes are described in terms of gas solubility coefficient, S , and diffusivity, D , and thus permeability, P , where $P = SD$ [18]. The solubility coefficient (hereafter referred to as solubility) is defined as the ratio of the radon concentration in a given compound to that of the surrounding air under equilibrium absorption conditions. The solubility, S , is a measure of the amount of radon absorbed by a given organic solid (or liquid) and is numerically equivalent to the concentration factor, the ratio of the concentration of radon in a given medium to the concentration of radon in air [19].

The radon solubility of air is used as the reference and is $S = 1$. In analogy with the radon sampling by liquid scintillators, new passive radon samplers have been developed by exploiting the radon absorption by different types of thermoplastics, which are characterized by a glass transition temperature, T_G , below which they are like liquids (rubbery), while above T_G they are like glasses. In particular, the plastics, used for Rn-sampling, are respectively silicone [20], polyethylene [2], polycarbonate [21-22], and polystyrene [23]. Among the plastics used to date, polycarbonate and polystyrene have glass transitions temperatures of about 100°C and 150°C respectively and can thus be considered glass-like. By contrast, polyethylene (PE) and polydimethylsiloxane (PDMS) have a T_G of -80°C and -150°C respectively, which are well below the room temperature and they are rubber-like- or liquid-like plastics. The permeability, P , is dominated by the diffusion coefficient D . Variation in D versus temperature are by far greater than those in S [24]. In the case of the glass-like plastics, such

as polycarbonate and/or polystyrene the radon solubility is expected to be little affected by temperature [3].

Among all the glassy polymers, studied to-date, the polycarbonate is the most attractive for radon sampling respectively because of its large glass transition temperature and its large Rn-solubility. Moreover, extensive investigations have been made in the past [25] about the temperature effects on the plastics, used as track detectors. According to these investigations, track fading occurred essentially when plastics were brought at temperatures above their T_G [26], i.e. for plastics in their rubber-like state. It was because of these unique characteristics that the polycarbonate has proved to be an excellent retrospective dosimeter [21-22]. Among the plastics listed in table 1, the CR-39 (Columbia Resin, 1939) is characterized by the lowest Rn-solubility, $S = 1$. The CR-39 detector is a thermoset plastic, the structure of which is knit together by cross-linking bonds, thus having little free volume and very low gas diffusivity [27]. Thanks to these characteristics and its large radiation sensitivity, the CR-39 is a very attractive track detector [27].

As it is clear from Table 1, the polycarbonate films are characterized by one of the largest radon solubility, while the CR-39 track detectors the lowest. Thanks to this combination of characteristics, it was possible to develop a simple radon badge by facing a polycarbonate film (used as radiator) against a CR-39 track detector [28]. In this radon film badge, the alpha particles from absorbed radon and its decay products may escape from the polycarbonate radiator thus penetrating the facing track detector, where they can be registered after a suitable chemical etching. The radon film badges are very similar and sometime identical to neutron film badges [29,30], which have been successfully used for personnel dosimetry throughout the world [27]. As reported in Table 1, the polycarbonate Rn-solubility can be by far larger than that of the widely used scintillation liquid: Opti-fluor [31]. Thanks to these characteristics and their thermal stability, the polycarbonate films can

Table 1. Characteristics of the plastics used for sampling radon by absorption processes.

	T_G (°C)	Solubility S
Air	1
Polydimethylsilicone -DMPS-(rubbery)	-120	-15
Polyethylene-PE (rubbery)	-80	~4
Polystyrene -PS (glassy)	100	20 ÷ 60
Polycarbonate-PC (glassy)	150	30 ÷ 100
CR39 (thermoset)	~1

successfully replace the organic scintillators for radon measurements in-water [3].

The film badge response, τ , under equilibrium-sorption conditions, can be considered to a good approximation given by:

$$\tau = k SCR \quad (2)$$

where k is a constant, S the radiator solubility, C the radon concentration to be measured and R the maximum range of the alpha particles within the radiator.

The radon film badge with the lowest response is formed by using a non-porous metal layer with $S = 0$. In this case, the response is due to the radon absorbed directly into CR-39 which has a solubility $S = 1$, i.e. a radon solubility equal to that of radon in air. As it can be derived from equation 2, the solubility of any compound can be evaluated by using it as radiator against a CR-39 and by evaluating its response to radon under equilibrium sorption conditions. More in general, the same approach can be developed for any-sorption-based radiator, after due corrections needed when using radiators with different densities.

4. Film Badges with Fast Response

In order to obtain a radon-film badge with fast response, it is important that the conditions of equilibrium absorption are achieved in a time much shorter than the mean radon-decay time-constant. When a gas diffuses through a membrane, there is a time lag, T , from the time the gas first enters the membrane until the steady state of flow is established. Using appropriate solutions of the diffusion equation, the time lag, T , can be related to the diffusion coefficient, D as:

$$T = d^2/6D \quad (3)$$

where d is its thickness of the membrane [28].

According to equation 3, by using a polycarbonate radiator (for which $D \sim 10^{-10} \text{cm}^2/\text{s}$) with a thickness of about 5 microns, it is possible to reach the equilibrium-type of radon absorption in a few hundreds of seconds, thus obtaining a radon film badge with uniquely fast response. Since the radiator thickness is by far less than the maximum range of alpha particles from the radon decay series (namely 64 microns), a radiator with an infinite thickness (i.e. thickness > 64 microns) can be achieved by using a stack of 13 films or more. For stack thicknesses less than 64 microns, it is possible to change the response sensitivity of the badge simply by changing the stack thickness. Since radon diffuses freely between any two-films interface, the time T to achieve the equilibrium absorption for a stack is the same of that of a single film [32]. The free in-air-diffusion of radon between

any two films of the stack is ensured by enclosing the badge in a permeation polyethylene bag, which drastically reduce the entry of water vapor. A radon film badge with a fast response can also be obtained by using a tissue layer made by plastic microfibers (long fibers with diameters less than about 10 microns), woven or knitted together to form a cloth. These microfibers may be suitably manufactured, for example, by cellulose derivatives, polyester, rayon, nylon, polyethylene, polypropylene, polyamide, other polymeric materials or combination thereof [33]. These clothes are characterized by a large surface to volume ratio, which makes it possible to achieve a fast equilibrium absorption.

By way of example, a cloth made of phenolic microfibers, known as Kynol [36], has been placed against a CR-39 as a radiator of the radon film-badge. The Rn solubility of this microfiber cloth has resulted to be $S=14 \pm 1$. Eventually, the most interesting development, pertinent to the radon film badges, was the discovery [35] that by heating the Kynol clothes in steam 700-900 °C, they could be transformed into a range of high surface area activated carbon cloths. The Activated Charcoal Cloth (ACC) is a unique form of carbon which is 100% pure activated charcoal in a textile configuration, with a high sorption capacity of gas and/or organic vapors. The surface to volume ratio of said ACCs is about two orders of magnitude larger than that of most activated-charcoal grains. Said large surface-to-volume ratio ensures a fast kinetic of adsorption and desorption of any gas (e.g. radon). These activated carbon cloths can be regenerated within 1 hour at 100°C in a ventilated oven. Incidentally, this regeneration procedure is the same for all the radiators for the radon film badges, listed above. Different types of activated carbon cloths have been used as film badges radiators [36]. As usual, the entry of water vapor has been limited by enclosing the film badge in a polyethylene permeation-bag. Radon film badges have been obtained by using different types of activated carbon cloths, derived respectively from the Kynol micro-fibers (Nippon Kynol Company) and from the cellulose-derivatives-based microfibers, known as Zorflex-FM1-250 (registered trade name by Charcoal Cloth International).

Table 2 reports the responses in terms of tracks/(cm². kBq.h/m³) of different types of radon film badges with fast-time response, obtained by using radiators made by thin plastics films and microfiber-based cloths. The solubility, S , of the Kynol microfiber changes from about 14 ± 1 into 2000 after the thermal activations. From this table is appear clear that radon film badges can have a response sensitivity which may differ for more than three orders of magnitude. By contrast, it is very hard if not impossible to change the response of the existing passive radon monitors developed for dwellings. Each radon film badge is enclosed in a heat-sealed polyethylene bag, which is characterized by a fast Rn-

Table 2. Radon film badges with similar fast responses but different response sensitivities.

Radiator	Solubility S	Response [Tracks.m ³ /(cm ² . kBq.h)]
Non porous aluminium	0	0,010 ± 0,002
Polycarbonate	80 ± 1	0, 80 ± 0, 10
Kynol cloth	14 ± 1	0, 14 ± 0, 01
Kynol 10 Activated carbon cloth	2000 ± 200	20 ± 2
Zorflex FM1-250 Activated carbon cloth	600 ± 60	6,0 ± 0,6

permeation-time, because of the film-badge small volume, as it can be derived from equation 1, However, in the case of Kynol 10, the equivalent volume of 1 cm² of 0.5mm-thick cloth is about 30 cm³ as it can be evaluated based on its large radon solubility, $S = 2000$. In order to keep these permeation bags airtight, it is necessary to protect them from any accidental hole. This is ensured by enclosing the permeation-based badge in a Tyvek bag, which is puncture resistant.

5. Radon Monitors with On/Off Response

The passive monitors, used for long term radon measurements are typically not equipped with a mechanism to turn them on/off, which is necessary to measure radon exposure during working hours only. The most popular radon monitors, capable to be turned on and/or off, are the electrets. These monitors are turned off by a shutter mechanism, which protects the electret sensor from the exposure to the radiation from radon and its decay products. When the exposure starts, the minimum response time of any radon monitor is about three hours, which is the time required to achieve the radioactive equilibrium between radon and its progeny. This is known as the “ramp up” time, during which there is a deficiency of response. By contrast, at the end of the exposure, provided that the radon diffuses rapidly from the monitor, the exposure to the radon progeny continues for three more hours, known as the “ramp down” time. This additional exposure compensates the initial response deficiency. By using a shutter to stop the detector response, there is no more this compensation and the response “deficiency” becomes unacceptable for the assessment of the 8-hours long (or less) workplace-exposures [37]. The best solution is to get rid of the shutter and to turn off the radon monitor by placing it in an environment with negligible radon concentrations [32]. For what concerns the end of the exposure, two possible errors may occur, due respectively to an accidental exposure in case the monitors are enclosed in a non-radon-proof bag, or to an additional exposure in case they are enclosed in a radon-proof bag, since the radon still present in the monitors

(and/or in their plastics components) continues the monitor exposures within the bag.

Finally, these problems can be solved by using a radon-free bag, which can be uniquely simple when designed for a Rn film-badge, because of its small sizes (namely less than 1 cm³). In this case, a radon free bag can be obtained simply by using about one gram of activated charcoal with an adsorption coefficient of about 4000cm³/g, enclosed in the bag. In practice, under equilibrium situation, one gram of activated charcoal adsorbs essentially all the radon i.e. it adsorbs all but 1/4000 of radon [38]. However, the key strategy is to avoid the use of activated charcoal, since, in addition to its handling difficulties, it requires a too long time to achieve the adsorption equilibrium. By contrast, the activate carbon fiber cloth, in addition to be very simple to handle, is characterized by a surface-to-volume ratio hundreds of time larger than that of activated charcoal, thus ensuring a fast equilibrium adsorption [36]. Moreover, this radon free bag is also very useful to drastically reduce the exposure of the monitors in case they are enclosed in a Rn-leaky bag. In this case, the radon entered into the bag will be essentially adsorbed all but a very small fractions, by the activated carbon fiber cloth. Incidentally, the transparent water-proof bags, used for smartphones, with special regards to those equipped with pressure-types of sealing, are of great interest as radon-free bag. As an important aside, these small transparent bags, in addition to be very strong and inexpensive, make it very ease to inspect the correct use of the personal radon dosimeter of each worker.

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