

Radon film-badges versus existing passive monitors based on track etch detectors

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Abstract. New methods and apparatus for the passive sampling of radon are introduced in the present paper. These samplers are based on the sorption of radon by layers of different types of solids. These solid layers result in sources of alpha, beta or gamma radiations, which can be detected by any passive or real-time detector, thus obtaining a variety of new radon monitors. By way of example, by facing one of said layers against a nuclear track detector, it is possible to obtain a compact radon film-badge. This film badge makes it finally possible to solve most of the shortcomings of existing passive monitors, based on track detectors, for the measurements of short- and long-term exposures of radon in air, in water, and in soils. These devices can be easily implemented by any existing radon service just as a back-up technology, with little or no change of the presently-used passive monitors.

Key words: radon • radon monitor • diffusion chamber • film badge • absorption • adsorption

Introduction

New methods and apparatus for the passive sampling of radon are introduced in the present paper. These samplers are based on the uptake of radon gas by different types of solids. In general, the uptake of any gas by solids has been termed “sorption” by McBain in 1909 [4] to include absorption and adsorption as special cases.

The absorption process considers that the sorption of a gas into a solid results in regions where the components form mixtures similar to actual solutions.

In the case of true adsorption, only the surface regions of the solid are considered to be involved in the sorption process.

Through a sufficiently large uptake of radon by absorption and/or adsorption processes, the solid-layers result in sources of alpha, beta or gamma radiations, which can be detected by any passive or real-time detector, thus obtaining a variety of new radon monitors.

By way of an example, by facing one of the said layers against a damage track detector, it is possible to obtain a compact radon film-badge [16, 17, 19], as described in the following. Parallel efforts have also been made to exploit these novel sorption samplers to measure radon by contamination monitors, such as pancake Geiger-Müller counters, which are typically not useful for the monitoring of radon and/or other gases [18].

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Table 1. Data of the partition coefficient and the diffusion constant of air, water, and two of the plastics used for alpha-track registration

Absorber	Thickness (μm)	Partition coefficient	Diffusion constant – D (cm^2/s)
Air	–	1	$\sim 0.1^{\text{a}}$
Water	–	0.25^{b}	$1.2 \times 10^{-5\text{a}}$
CR-39	1	0.06^{c}	–
Polycarbonate	0.3	8^{c}	$\sim 0.6 \times 10^{-10\text{d}}$

^a From Tanner [13].

^b The partition coefficient of water equal to the Osvald coefficient [2].

^c Value derived from More and Hubbard [5].

^d Value at 18.5°C derived from Pressyanov *et al.* [9].

Radon film-badges based on absorptive radiators

The processes of radon absorption in solid polymers have been already investigated in the past with a specific goal in mind to develop new radon monitors [3, 8, 9, 11, 14].

All the components of existing passive radon monitors are made of plastics, such as respectively the diffusive-chamber housings and the alpha detectors (track detectors and/or electrets). A thorough investigation of the radon absorption by plastic components used for passive monitors has been made by Moore and Hubbard [6]. Table 1 reports the data derived from these investigations, such as respectively the absorptive-film thickness, radon diffusion, D , and radon partition coefficient (related to air) of some of these plastics. In particular, similar data concerning air and water are also included in Table 1.

The partition coefficient is a measure of the absorption phenomenon and is given by the ratio of the radon concentration in the absorption material and that in the air. Under non-equilibrium absorption, as in the case of the plastics listed in Table 1, the said partition coefficient is dependent on the plastics thickness. Only under steady state conditions (i.e. equilibrium absorption) this partition coefficient becomes a characteristic of a given plastic, known as solubility [2]. The solubility of organic solids has been first exploited for both radon- and helium-measurements by Guerin and Vuilleminot since 1995 [3]. In the case of organic liquids, the radon solubility has been exploited for radon measurements since the early 1980's [10].

From the partition coefficients listed in Table 1, it appears clear that the CR-39 is of interest as a detector for the radon film badge, while polycarbonate is more suitable as a radiator.

However, Pressyanov *et al.* [8, 9] have developed an ingenious retrospective radon dosimeter by exploiting a thick polycarbonate basis, used for compact disks (CDs). In this case, the polycarbonate is both absorptive radiator and track detector.

According to the list of Table 1, the diffusion constant of radon in polycarbonate is about one billion times smaller than that of radon in air. For these reasons, the equilibrium conditions for in-air radon-diffusion into existing passive chambers are rapidly achieved, while the equilibrium conditions for radon absorption into polycarbonate can hardly be achieved in a time much smaller 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

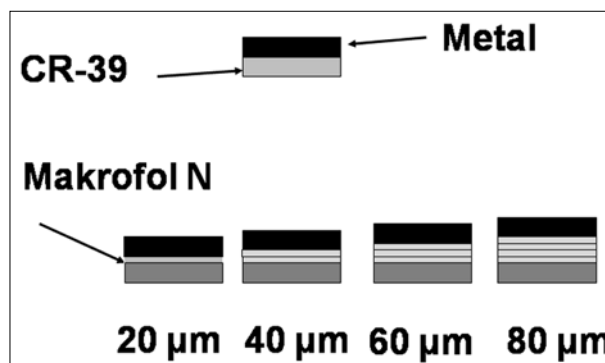


Fig. 1. Cross-sectional view of different radon-badges.

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 in the following [1]:

$$(1) \quad T = d^2/6D$$

where D is the radon diffusion in the membrane in unit of cm^2/s and d is its thickness in cm.

In order to obtain a fast response-time for the radon badges (i.e. a time T much smaller than the mean radon-decay time constant, τ), it is necessary to use a radiator with sufficiently small thickness, d , as it can be derived from Eq. (1). Actually, Eq. (1) is based on the uptake of radon from only one surface, so that it can be considered valid only for film thickness much larger than the radon diffusion length, which is given by $(D\tau)^{1/2}$. For films with thickness less than the said diffusion length, the time lag, T , evaluated according the Eq. (1) is obviously larger than the true value.

With a 20 μm thick polycarbonate film (Makrofol N from Bayer AG), the mean time to achieve radon absorption under equilibrium conditions is less than 3 h, which is by far smaller than the mean radon decay time constant, τ (132 h).

Figure 1 reports a schematic view of different radon film badges obtained by using one or more radiators of 20 μm thick film of polycarbonate (Makrofol N from Bayer AG). Since radon diffuses freely between any two film interfaces [7], the time T to achieve the equilibrium absorption for a stack of radiators is the same as that of a single radiator. Table 2 shows the response of the radon film badge vs. the radiator thickness. This response reaches a plateau value at about 60 μm , which can be explained considering that the maximum range in polycarbonate of the alpha particles involved is 64 μm .

Table 2. Response of radon film badges obtained by using one or more radiators of 20 μm thick film of polycarbonate (Makrofol N from Bayer AG)

Radiator	Radiator thickness (μm)	Response [Tracks·m ³ /(cm ² ·kBq·h)]
Non-porous aluminium sheet	100	0.01 \pm 0.002
1 Makrofol N	20	0.20 \pm 0.02
2 Makrofol N	40	0.30 \pm 0.02
3 Makrofol N	60	0.41 \pm 0.04
4 Makrofol N	80	0.40 \pm 0.05

When the thickness of the radiator is less than the maximum range of the alpha particles involved, it is important to add a back-up support with negligible radon absorption, as, for example, a metal sheet.

When this sheet (such as a non-porous aluminium sheet) is placed against the CR-39 with no absorptive radiator in between, the lowest response sensitivity is obtained, which is determined by the radon absorbed into the CR-39 detector itself. This response can be considered as the intrinsic response of CR-39.

For purely accidental reasons, the ratio between the maximum- and the minimum-response of Table 2 is equal to 40 ± 5 , which is equal to the radon partition coefficient between polycarbonate and air, under conditions of equilibrium absorption [6]. As stated above, under these conditions and/or when the time to achieve the equilibrium, T , is much less than the mean radon-decay time constant, the partition coefficient becomes equal to the radon solubility of the polycarbonate.

The largest value of the radon solubility for polycarbonate, reported recently by Pressyanov [8], is 26.4 ± 2.5 , which is smaller than the value reported above. This difference may be due to the different morphological structures of the polycarbonate films used.

The Makrofol N has been termed CR-40 for being a carbonate radiator which makes it possible to increase the intrinsic response of CR-39 by a factor of 40. Alternatively, it is possible to state that under equilibrium absorption the radon-concentration of the CR-40 radiator is 40 times of that of the surrounding air.

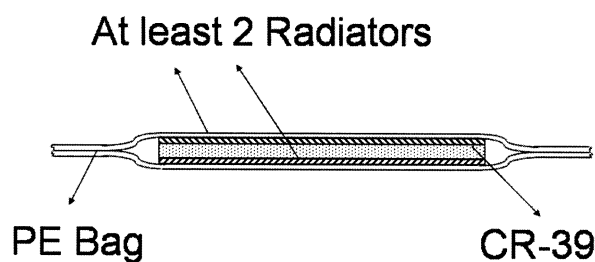
An extensive analysis has been carried out recently which made it possible to identify plastic radiators capable of increasing the intrinsic CR-39 response sensitivity by a factor up to about 100.

Radon film badges with two different responses

Radon film badges make it possible to obtain a compact radon monitor with at least two different response sensitivities.

Figure 2 is a vertical cross-section view of a radon film badge, designed to enable the measurement of large radon exposures with two different response sensitivities, thanks to the use of the two absorptive radiators, facing the opposite surfaces of the CR-39 track detector.

Alternatively, instead of facing the two opposite surfaces of the track detector, the said radiators can be placed side by side on only one of the surfaces of the track detector. The number of responses of the radon film badge, shown in Fig. 2, depends on the number of radiators used.

**Fig. 2.** Radon film badge with two absorptive radiators enclosed in a heat-sealed polyethylene bag.

In case only one radiator is used facing only one of the detector surfaces, the opposite surface would always register alpha particles escaping from any material, which comes in contact with the said surface. As already stated above, even if the material facing the said detector-surface is a non-porous metal with no radon absorption, the mentioned surface will still register tracks, due to the radon absorbed in the track detector itself.

In practice, the radon badge of Fig. 2 is characterized by at least two different response sensitivities.

The linearity of the response of the said passive monitor can cover a wide range of radon exposures provided that the absorptive materials are properly chosen.

In Fig. 2, each radiator is simply illustrated by a single layer of material. Actually, the radiator may be formed by one or more absorptive films, which are backed by a sheet of non-porous metal when their thickness is less than the alpha-particle range. As already stated above, the use of radiators with different overlapped films is made possible thanks to the free in-air-diffusion of radon within any two-films interface.

However, in the case of large relative humidity, the radon may not diffuse freely anymore within the said interfaces, since they could be filled with condensed water, characterized by radon diffusion coefficient, D , which is 10 000 smaller than that of air (see Table 2), thus causing a drastical increase of the response time.

This problem of humidity can be solved by enclosing the radon badge in a heat-sealed polyethylene bag (as shown in Fig. 2), characterized by a large permeability to radon and a small permeability to water vapour [15]. The most important requirement for this bag is that the permeation-time of the radon into the bag must be always much smaller than the mean radon-decay time-constant. The lower part of Fig. 3 shows the radon badge which has been schematically described in Fig. 2. This badge may be characterized by two-responses sensitivities, which could be just the same of those of the passive chambers, shown in the upper part of Fig. 3, developed respectively by Tommasino *et al.* [15] and by Urban and Piesch [20].

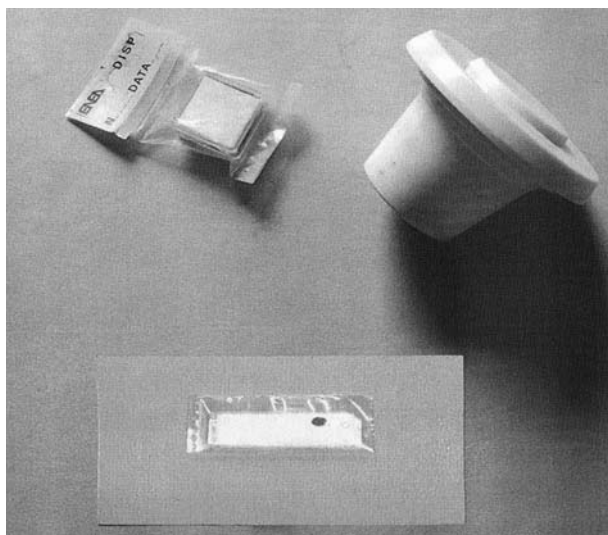


Fig. 3. Lower side: radon-badge with two different responses; upper side: two passive diffusion-chambers respectively from Tommasino *et al.* [15] on the left and from Urban and Piesch [20] on the right.

Radon-in-soil measurements with the diffusion chambers, require large holes in the ground, which are often conducive to wrong measurements, because of the large time required to achieve equilibrium diffusion of radon into the large holes, especially under the conditions of large relative humidity [13]. The radon film badge shown in the lower part of Fig. 3 is uniquely compact, thus requiring holes in soils with volumes thousands of times smaller than those needed for diffusion chambers.

This radon badge, once enclosed in a heat sealed polyethylene bag, makes it finally possible to carry out passive radon-measurements directly in water.

Radon film badges with adsorptive radiators

Passive diffusion chambers based on track detectors are not sufficiently sensitive for one-week indoor measurements.

A passive device based on track registration with a sufficiently large response for measurements has been reported by Sutej *et al.* [12], who proposed to use a track detector adjacent to activated charcoal.

One of the shortcomings of this device is that its response sensitivity may depend on the particular time of the exposure, specially because the activated charcoal is used as a non-equilibrium-type sampler. Equilibrium-type of sampling by activated carbon adjacent to a track detector has been obtained by van Deynse *et al.* [21], who have been using thin layers of activated carbon.

Unfortunately, these thin layers of activated carbon still present several shortcomings such as large cost, practical difficulties due to their grain-type morphology, and humidity-dependent adsorption characteristics.

Through extensive investigations, it was finally possible to identify adsorptive radiators with surface-to-volume ratios hundreds of times larger than those of activated charcoal, thus ensuring a fast time for achieving equilibrium adsorption.

In particular, an adsorptive radiator (ACF 1000 from Kareray Chemical Co. Ltd.) has been chosen for giving

the desired response. Following the terminology used for the absorptive films, this radiator has been termed CR-2000, since it increases the intrinsic response of the CR-39 by a factor of 2000. Alternatively, it is possible to state that the radon-concentration of the CR-2000 radiator under equilibrium adsorption is 2000 times of that of the surrounding air.

It is expected that any radon badge formed by an adsorptive radiator facing a track detector would have a response highly dependent on humidity. As already suggested above, this humidity problem can be solved by enclosing the adsorptive badge in a heat-sealed polyethylene bag [15].

The use of this permeation bag requires a careful choice of all the parameters involved (such as the quantity of the absorptive material, surface area, volume and thickness of the bag) in order to ensure that the equilibrium sampling is achieved in a total time, which is much smaller than the mean decay-time constant of radon.

By using a CR-2000 radiator with a thickness of about 2.5×10^{-2} g/cm² the response is:

$$(2) \quad R = (20 \pm 4) \text{ tracks} \cdot \text{m}^3 / (\text{cm}^2 \cdot \text{kBq} \cdot \text{h})$$

This response is more than 20 times larger than those of the passive chambers shown in the upper part of Fig. 3. This large-response badge greatly facilitates radon-measurements for short-term (from a few days to one week) exposures indoors by passive monitors based on track-etch detectors.

Conclusions

Radon film-badges with any desired response can be obtained with radon-sorption radiators. By using the CR-39 track detector, this response can be easily changed within a range of three orders of magnitude, i.e. from a response of 0.01 (tracks·m³/(cm²·kBq·h)) with a metal radiator to 20 (tracks·m³/(cm²·kBq·h)) with the CR-2000 radiator.

When compared with the existing passive radon monitors based on track-etch detectors, these radon film-badges present several advantages, such as unique compactness, wide linearity of response, any desired response sensitivity, possibility to carry directly-in-water radon measurements.

All the investigations described above have been carried out by using different types of sorption radiators facing track detectors. It can be safely anticipated that these radiators can be successfully used also with other detectors of alpha particles, such as electrets, thermoluminescent materials, silicon diodes, etc.

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