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To cite this version:

HAL Id: jpa-00246295
https://hal.science/jpa-00246295
Submitted on 1 Jan 1990

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Automated radon monitoring system for continuous environmental sampling

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(Received 30 April 1990, revised 9 August 1990, accepted 7 September 1990)

Résumé. — On a développé un appareil pour la mesure en continu de la concentration du radon dans l’air. Le fonctionnement se base sur la capture électrostatique des produits de $^{222}$Rn (ou $^{220}$Rn) et sur l’observation de la désintégration $\alpha$ du $^{218}$Po et $^{214}$Po (ou $^{212}$Bi et $^{212}$Po pour $^{220}$Rn). L’appareil se montre particulièrement indiqué pour la mesure courante dans des environnements fermés tels que résidences et immeubles.

Abstract. — An automatic instrument for the continuous measurement of radon concentration in air has been developed. The system operates by electrostatically collecting the daughter products of $^{222}$Rn (or $^{220}$Rn) and observing the $\alpha$ decay events from $^{218}$Po and $^{214}$Po (or $^{212}$Bi and $^{212}$Po for $^{220}$Rn). The detector is particularly well suited for routine measurements in enclosed environments such as residences and other buildings.

Introduction.

For a long time radon emanation in the earth and in the atmosphere has been the subject of numerous studies, but, only recently, the presence of radon in air is considered with increasing attention; in fact owing to the restricted exchange of air in closed environments the gas may achieve levels dangerous for human health.

$^{222}$Rn, an inert gas, is part of the $^{238}$U decay series. Its immediate parent, $^{226}$Ra, is an ubiquitous constituent of crustal or subcrustal materials, and is found in soils and in building materials having rock- and soil-based components. When the alpha-decay of $^{226}$Ra produces $^{222}$Rn, the newly formed nucleus assumes the properties of a gas. Radon gas, escaping from the ground and building materials, may enter the houses through cracks, drains, basements. $^{220}$Rn (thoron) may also be present in indoor air but, in general, its concentration is limited by relative short (55 s) half-life [1, 2].

These radon (or thoron) decay products are chemically active and can attach to surfaces, such as room walls, airborne particles, or upon inhalation, lung tissue. This latter process is primarily responsible for the health effects associated with radon.

The need to measure radon at low levels for extended periods with low maintenance, requires the development of a special detector.

In this work we present an automated instrument for measuring daily fluctuations of $^{222}$Rn (or $^{220}$Rn) concentration inside closed environments. Some experimental tests in different sites have been effected.

The measurement process.

Short-lived radioactive daughter products of $^{222}$Rn and $^{220}$Rn, obtained by $\alpha$ decay of their respective parents, behave as small positive ions (by the stripping of orbital electrons due to $\alpha$-particle emission and/or to atomic recoil mechanisms). Also $^{222}$Rn and $^{220}$Rn progeny obtained by $\beta$ emission is preferentially formed in a positively charged state. However, $^{222}$Rn (and $^{220}$Rn) progeny may be also found in a negatively charged or electrically neutral state, but this progeny constitutes a relatively small fraction of the total [3-7].

The fact that $^{222}$Rn and $^{220}$Rn progeny is initially formed in an electrically charged state suggests the possibility of using electrostatic collection of the charged decay products for a measurement technique.
The figure 1 shows the basic architecture of the instrument.

The sampler of radon consists of a hemisphere (~ 4 dm$^3$) covered with 2.5 cm of open-pore polyurethane foam.

The hemisphere is connected to ~ 3 500 [V] with respect to a commercially available silicon diffused junction detector (EG & G ORTEC Model N` R-024-450-100) located at the centre of the hemisphere.

Radon, but not its daughters, diffuses through the porous foam filter and enters a sensitive volume. The degree of passive diffusion is determined by the differential radon concentration gradient inside and outside the detector.

The diffusion coefficient of radon in our open pore foam is 0.032 [cm$^2$/s] [1, 7]; about 30 minutes are required for establishing (90%) by passive diffusion equilibrium inside the sensitive volume.

For each $^{222}\text{Rn}$ atom that decays within the sensitive volume is formed a $^{218}\text{Po}$ (RaA) (~ 88% of the time the RaA is formed as a positive ion) [7].

The electrostatic field applied within the sensitive volume collects the $^{218}\text{Po}$ (positive ions) on the surface of the detector. The RaA attached on the surface of the detector decays in $^{214}\text{Pb}$ (RaB) by $\alpha$ (6.0 MeV) emission. Subsequently RaB decays in $^{214}\text{Bi}$ (RaC), and $^{214}\text{Po}$ (RaC'). The $^{214}\text{Po}$ decays in $^{214}\text{Pb}$ (RaD) by $\alpha$ (7.69 MeV) emission, as shown in the scheme of figure 2a.

Thus, for each $^{222}\text{Rn}$ atom that decays within the sensitive volume, two $\alpha$ particles are potentially observed.

When $\alpha$ radiation reacts in the detector, negative pulses are generated. These pulses are amplified by the preamplifier located at the bottom of the detector.

The output pulse of the preamplifier is sent, after shaping and filtering, to a Single-Channel Analyzer (SCA). The SCA produces a logic output pulse (TTL) only when the preamplifier output pulse falls within the energy window set by $E$ and $\Delta E$ control. Thus a particular spectral line may be selected and counted.

The figure 3 shows the $\alpha$-spectrum observed by our instrument and visualized with an external multichannel analyzer.

There is a difference in the ratio of two peaks. The peak of the RaA is lower than that one of the RaC'. Since each RaC' atom is produced from a RaA atom, the lower collection efficiency of RaA indicates a loss mechanism, due probably to a recombination of $^{218}\text{Po}$ positively charged.

The exact mechanisms for $^{218}\text{Po}$ neutralization are not well known; different and even contradictory results have been obtained under various conditions; however the neutralization of Polonium — 218 ions is a function of the physical and chemical properties of the ambient air, such as described in literature [3, 4, 7], while the collection efficiency of RaC' appears to be unaffected by normal fluctuations of relative
humidity. The study of loss mechanisms in different environmental conditions will be object of a further research.

RaA in a negatively charged or neutral state is not collected on the surface of the detector; on the contrary RaB, RaC and RaC', which behave as positive ions, are collected.

In this way it is possible to detect the decay of RaC' without detecting RaA.

For this reason it is used the peak region of the RaC' for selecting the levels of the SCA.

For the $^{220}$Rn measurement we selected the peak of the $^{212}$Po (ThC'). The thoron decay chain is shown in figure 2b.

The instrument may record simultaneously the $^{222}$Rn and $^{220}$Rn concentrations by utilizing two SCA (the levels of two SCA are selected using the peak region of the RaC' and the ThC').
The sampler is constructed around a microprocessor with a structure suitable for managing controlling units and for doing arithmetic operations. Through a proper firmware the processor controls all the operations as well as the measuring procedures of the sampler.

A second microprocessor, identical to the first, is installed to control the data communications in the air pollution monitoring networks.

The two microprocessors are connected by a Dual Port - Random Access Memory. This particular device allows us to obtain an interchange of data without dead time in the measurement system.

Figure 4 shows the complete radon monitoring system.

**Measurements.**

The instrument was calibrated by exposing the sensitive volume to various known concentrations of radon, using the technique described in references [8, 9] and observing the equilibrium response of the detector. Several exposures to different concentrations of $^{222}$Rn were made to insure that the detector response is linear with concentration.

A calibration and background determination were made after the instrument underwent extensive field operation (about six months) in the gallery of Gran Sasso Laboratory.

The calibration factor remained a constant value of 0.76 counts per minute for a radon concentration of 1 Bq/m$^3$ and the instrument background is 0.04 counts per hour.

The minimum significant measurable activity [10] in 60 minutes counting period, at the 95% confidence level, is 0.012 Bq/m$^3$.

The response time of the monitor was determined by observing the count rate, as radon was initially injected into the test chamber. A steady state condition between the radon concentration in the atmosphere and the sensitive volume was achieved within 30 minutes. There is a time lag between a change in the ambient radon concentration and the instrument response which is related to the diffusion time and the time necessary for buildup of the short-lived daughters on the detector.

Measurements of indoor radon concentration were made in a ventilate room and in the gallery of Gran Sasso Laboratory (2000 meters underground). The data set demonstrated a difference in the radon concentration between University of L'Aquila Laboratory (Fig. 5a) and Gallery of the Gran Sasso Laboratory (Fig. 5b). In the natural gallery, where the humidity and temperature are constant, the radon emanation shows a typical regular trend, while in a residential room of average ventilation the...
environmental parameters seem to influence the daily radon fluctuation and its level.

Conclusions.

The main objective was to develop a new instrument for continuous automatic monitoring of natural concentrations of $^{222}\text{Rn}$ (or $^{220}\text{Rn}$) in indoor environments.

The measuring procedure provides not only a value for mean concentration of $^{222}\text{Rn}$, but also indicates the variation of the concentration over the measuring period.

The system was designed without the use of air movers or pumps. The unit is unobtrusive and acceptable into a daily routine without disrupting business activities.

The presence of two microprocessors, in the architecture of the instrument, makes the sampler well suited for incorporation into pollution monitoring networks.

The sensitivity of the instrument is comparable with that of other systems commercially available; its effective cost is low and convenient ($\approx 3000\$$).

The high flexibility and reliability in operation and transmission of data was checked in early uses of the instrumentation.

Acknowledgments.

We would like to acknowledge the S.E.A. (Strumentazione Elettronica Avanzata Via Tiburtina, 1010 00156 Roma Italy) for support in instrument realization.

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