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# *Metrology of the Radon in Air Volume Activity at the Italian Radon Reference Chamber*

*G.Sciocchetti, G.Cotellessa, E. Soldano, M. Pagliari*
*Istituto Nazionale di Metrologia delle Radiazioni Ionizzanti*
*ENEA Centro Ricerche Casaccia*
*C.P 2400 I-00100 Roma*
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*Full postal address of all co-authors:*
*ENEA C.R. Casaccia, Via Anguillarese, 301, 00060 S. Maria di Galeria Roma (Italy)*

*<u>Abstract</u>*

*The approach of the Italian National Institute of Ionising Radiations (INMRI-ENEA) on radon metrology has been based on a complete and integrated system which can be used to calibrate the main types of $^{222}Rn$ in air measuring instruments with international traceability. The Italian radon reference chamber is a research and calibration facility developed at the Casaccia Research Center in Roma. This facility has an inner volume of one $m^3$. The wall is a cylindrical stainless steel vessel coupled with an automated climate apparatus operated both at steady and dynamic conditions. The control and data acquisition equipment is based on Radotron system, developed to automate the multitasking management of different sets of radon monitors and climatic sensors. A novel approach for testing passive radon monitors with an alpha track detector exposure standard has been developed. It is based on the direct measurement of radon exposure with a set of passive integrating monitors based on the new ENEA piston radon exposure meter. This paper describes the methodological approach on radon metrology, the status-of-art of experimental apparatus and the standardization procedures.*

## *Introduction*

*In Italy, the metrological activity is carried out under the law act N. 273/1991 concerning the Organisation of the National System of Calibration. This act recognised ENEA the Italian National Metrology Institute of Ionising Radiation (INMRI) for the reference standards of units in the field of ionising radiation. The INMRI-ENEA is operating a radon quality system that includes a traceability chair to national and international calibration standards. This paper describes the methodological approach on radon metrology and the state-of-the-art of experimental apparatus developed at the ENEA Centro Ricerche Casaccia in Roma. The approach of ENEA-INMRI on radon metrology has been based on a complete and integrated system which can be used to calibrate the main types of $^{222}Rn$ in air measuring instruments with international traceability. This system comprises primary and secondary standards for $^{226}Ra$ and $^{222}Rn$ activity, transfer standards, and reference instruments.*

*Exposure chambers have been developed to set up radon standard atmospheres, traceable to national standards, used for the calibration of radon and radon progeny measuring instruments. The INMRI-ENEA climate chamber was designed to set up environmental conditions for the calibration of radon and radon progeny measuring instruments. The test facility described in this paper provide controlled environment needed for instruments and methods evaluation. The unique characteristics of the INMRI-ENEA climate technology of the test chamber allow fully automated conditioning cycles, that doesn't influence the aerosol concentration and size distribution of reference atmosphere. The exposure of measuring devices to known $^{222}$Rn concentrations in a radon chamber provides the best conditions for the calibration of continuous and integrating $^{222}$Rn and radon progeny monitors. Climate radon chambers enable controlled conditions in terms of temperature, humidity, air exchange-rate, radon concentration, aerosol size distribution, for conducting experimental tests on radon and radon progeny measuring equipment.*

*The $^{222}$Rn Reference Measurement System (RMS)*

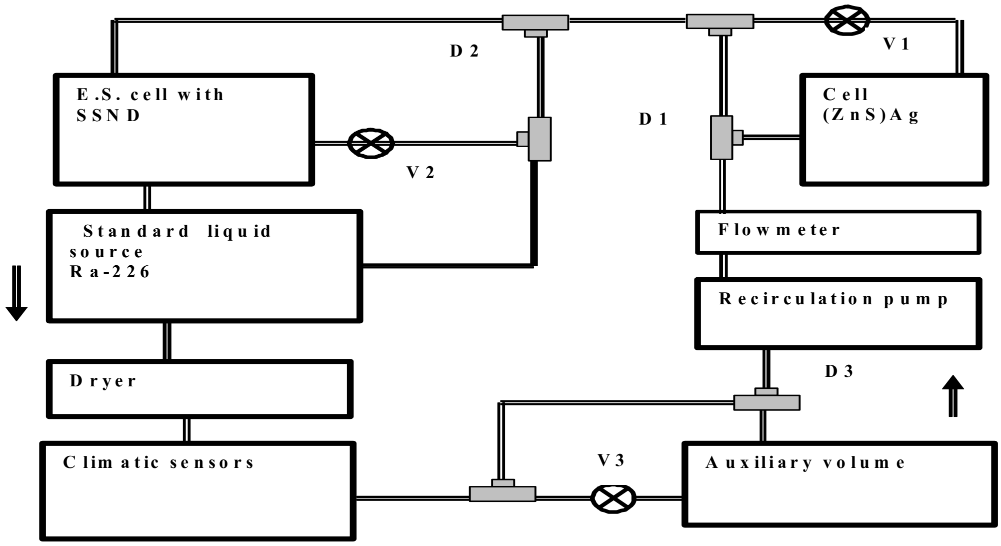


*Fig. 1 $^{222}$Rn Reference Measurement System (RMS)*

*The $^{222}$Rn Reference Measurement System (RMS) is an essential part of the INMRI-ENEA radon metrology system. It is used to calibrate $^{222}$Rn samples in glass bulbs or reservoirs of any geometry and volume. The concept of a radon reference standard based on the measurement method of $^{222}$Rn activity with the closed circuit technique was developed at the Radon Laboratory of INMRI-ENEA (1). The experimental apparatus is based on the closed circuit technique with the use of a recirculation apparatus and a flow-through radon measuring equipment. It is made of a radioactive source section, a gas circulation and dehumidification unit, a mass flow-meter and a flow-through $^{222}$Rn measuring instrument (2,3). The radioactive source section consists of calibrated $^{226}$Rn liquid sources in glass bulbs, traceable to the NIST. The $^{222}$Rn measuring instrument consists of a flow-through real-time monitor based on the alpha spectrometric measurement of $^{222}$Rn decay products electrostatically collected on the surface of a solid state detector. Due to its high spectral resolution it yields very precise measurements. The closed circuit, when calibrated with* *standard $^{226}$Rn liquid sources can be used as a measuring apparatus of radon samples in glass bulbs or in other containers. A procedure based on the measurement of radon gas with a calibrated closed circuit has been developed to calibrate any flow-through radon measuring instrument (self-calibration technique). The $^{222}$Rn Measurement System allows to calibrate the $^{222}$Rn Reference Instrument with traceability to the national $^{226}$Rn primary standard.*

*Apparatus used for the standardization of radon reference atmospheres: the Radotron system.*

*The Radon Reference Instrument (RRI) used for the standardization of the INMRI-ENEA radon test chambers is based on the Radotron system. The schematic diagram is shown in Fig.2. Radotron is a new concept of multifunction system designed and realised by ENEA with the financial support of an EU project. Radotron is operated running a custom LabView virtual instrument software to automate data-acquisition and evaluation. The apparatus based on Radotron system automates testing procedures and increases the accuracy of standardization of radon atmospheres. Moreover, it provides control functions with the capability to correlate data from different radon monitors and climatic sensors.*

*The test facilities are operated with radon measuring instruments connected through a recirculation circuit to the chamber, for sampling on-line the standard atmosphere. The equipment consists of two flow-through monitors working in the external circulation system: the reference instrument based on the spectrometric detection of the Rn decay products collected electrostatically on SSND detector and a backup total alpha counter based on scintillation technique. The spectrometric instrument provides readings every ten minutes and measures the radon volume activity with a very high precision, usually better, than the precision of the device which is being exposed within the chamber. The reading of the reference instrument and the system under test are carried out simultaneously for the evaluation of the calibration factor. All monitored data are automatically transferred to a database for later calculation of exposures and environmental conditions during each experimental run.*

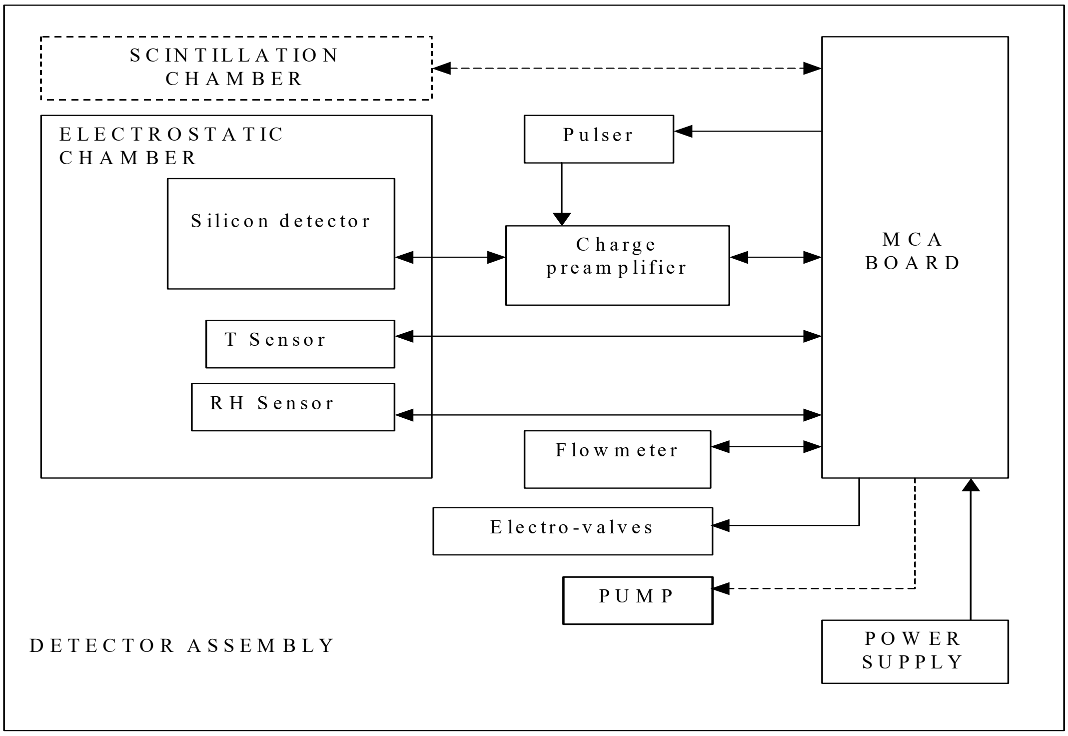


*Fig.2 - Schematic of Radotron system*

***The INMRI-ENEA climate radon chamber***

***The radon chamber is an experimental facility at the ENEA Centro Ricerche Casaccia for the development of new measuring techniques and accurate calibration procedures traceable to recognized radon standards of national and international institutes. The figure 3 shows the diagram of INMRI-ENEA climate radon chamber and the figure 4 the frontal view of the experimental apparatus.***

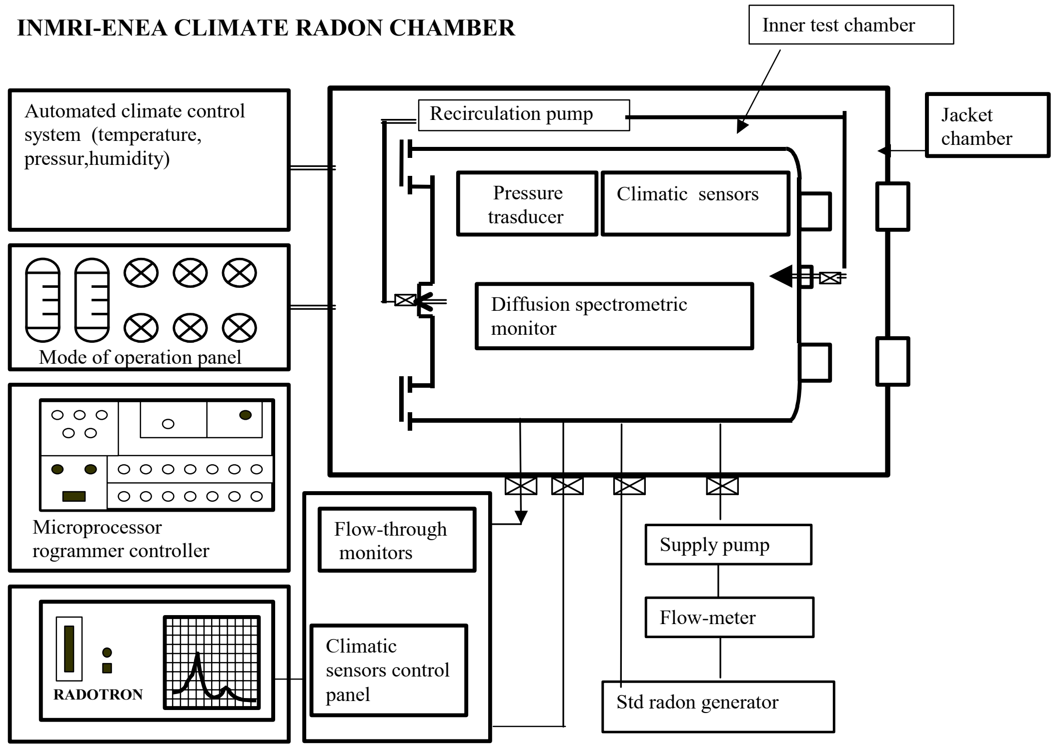


*Fig 3 Diagram of INMRI-ENEA climate radon chamber*

*Fig 4 Frontal view of the INMRI-ENEA Climate Radon Chamber*

*. Outstanding features of this apparatus are:*

- *unique characteristics of climate technology based on fully automated conditioning cycles of reference atmosphere that doesn't influence the aerosol concentration and size distribution*
- *automated monitoring of both radioactive and climatic parameters based on the new multifunction instrument Radotron*
- *self-calibrating measuring apparatus with traceability to the radon standard with the highest level of accuracy*

*This facility features a double wall design. The inner test chamber is surrounded by a jacket chamber which serves as a thermostat. The test chamber consists of a cylindrical stainless steel vessel of one cubic meter coupled with a climate control system. The jacket chamber is made of insulating elements laminated with metal sheets. The door is fitted with one window for illumination and two auxiliary openings. The facility is equipped with several sampling ports distributed over the two side walls. The special designed and constructed climate system allows to control the environmental parameters. It works at air-tight computer controlled temperature, pressure and humidity conditions. The chamber can be supplied with clean "aged" air or with dried and pre-filtered outdoor air. The air can be circulated inside and outside the inner vessel and heated, cooled, dried and moistened, without influencing the radon concentration of the confined atmosphere. The chamber is operated under static and dynamic conditions (4,5). Under the dynamic recirculation conditions the air mass is taken from the front side of the chamber and reintroduced through the rear wall. Under the dynamic open circuit conditions, the air mass taken from the chamber is continuously exhausted outside the internal useful volume. To ensure suitable radon homogeneity a ventilation fan is operated with radial air motion inside the cylindrical volume of the test vessel. Temperature can be varied over the range of -35 to 60 °C, pressure over the range of 700 to 1100 mBar and humidity over the range of 10 to 95 % (over temperature range of +5 to +60 °C , with + 4°C dew point limitation).*

*The chamber is in use for the calibration of active and passive radon measuring instruments. The figure 3 shows a schematic of the experimental apparatus used to standardize the reference atmosphere of the radon chamber. The front view of the experimental apparatus is shown in Fig.4. The monitoring techniques used for standardization purposes are based on devices integrated into the chamber and on air sampling external devices for measuring radiation and climatic parameters. The basic experimental apparatus consists of a radon generator and two real time instruments which measure radon concentration inside the radon chamber. Auxiliary instruments include two pumps, two flow meters and a system of climatic sensors for the measurement of pressure, temperature and relative humidity. The radon generator is based on a circulation system, which includes a small pump, a flow meter and $^{226}$Ra flow-through dry solid source. Bubbler with $^{226}$Ra calibrated liquid solutions, traceable to national standards are used for particular applications, e.g. for the direct calibration of chamber atmosphere.*

*The climate control provides stable conditions and variations over a wide interval of the environmental parameters, e.g. temperature, humidity, air pressure and aerosol concentration. This facility provides the best conditions to set up testing procedures of instrument performances based on the control of influence quantities, e.g. climate and aerosol parameters (concentration and size distribution). In particular the chamber allows to calibrate instruments with the output signal depending from environmental influence quantities.*

*The exposure chamber is operated with two standardization protocols (4). In the recirculation mode (closed circuit conditions), the output air from the exposure chamber was recirculated through the radon generator, at* *equilibrium with a $^{226}$Ra reference source (solid or liquid). Once the radon concentration is set to a particular value, there is control of the radon source and chamber parameters to maintain the radon level over the exposure period for the particular device being tested. In the open circuit mode the experimental conditions are established balancing the radon activity from the radon generator with the air change rate with the use of an extractor. Under the dynamic open circuit conditions, radon activity is flushed off from the $^{226}$Ra flow-through solid source and continuously exhausted outside the internal useful volume of the chamber. The increase in radon concentration in the chamber starts at the moment the pump of the radon generator is activated and stops when the radon balance is achieved inside the test chamber. The final radon concentration depends on the $^{226}$Ra source activity and the chamber volume. When instruments to be calibrated are set up in the chamber, the atmosphere is continuously sampled and measured using reference instruments. The radon atmosphere can be varied (and held stable) over the range of about 200 Bq $m^{-3}$ to about 100,000 Bq $m^{-3}$, by continuously flushing off the radon accumulated in $^{226}$Rn dry solid sources supplied from Pylon Company.*

*The radon exposure standard based on a system of alpha track detectors for testing passive radon monitors.*


*Fig. 5 ENEA Piston Radon Dosimeter (PRD)*

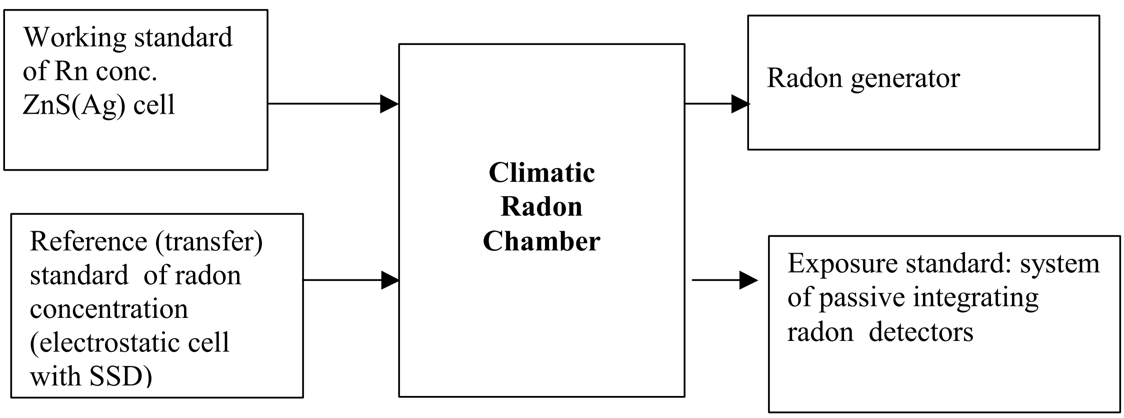

*Fig. 6. Schematic of the new experimental facility based on the PRD exposure standard at ENEA C.R. Casaccia.*

*The calibration of radon measuring instruments is usually carried out inside test chambers at well defined and stable radon concentrations. The response of a passive integrating detector must be compared with the "target value" of the time integrated radon concentration of the test atmosphere to which the device is exposed. The standardization of reference atmosphere is carried out with accurate measurement of radon concentration based on grab sampling or real time instruments.*

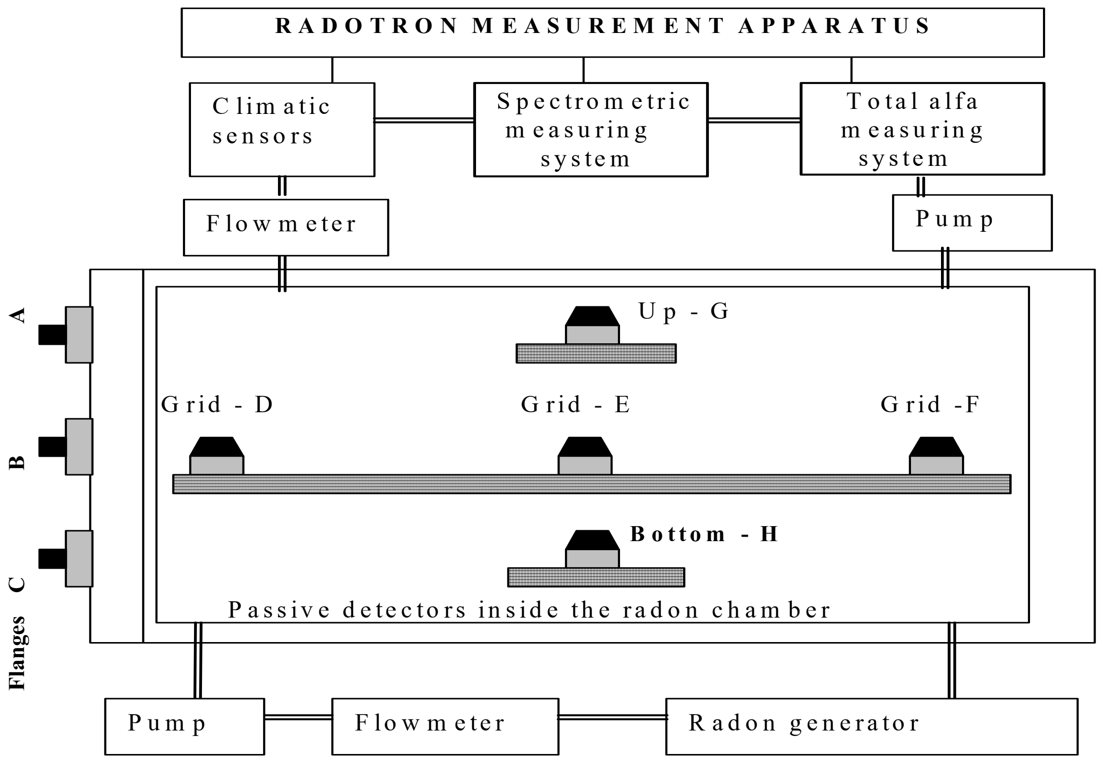


*Fig. 7. Experimental apparatus with the lay-out of RPDs used to test the exposure uniformity inside the radon chamber*

*A novel approach for testing passive radon monitors has been developed. It is based on the direct measurement of radon exposure with a set of passive integrating monitors based on the new ENEA piston radon exposure meter. A Radon Exposure Standard (RES) has been realized with a measuring apparatus based on a set of passive integrating monitors. The monitors are based on a new concept of passive time-integrating alpha track detector developed at the INMRI-ENEA laboratory. The novel device, called Piston Radon Dosimeter (PRD), uses a CR-39 detector fixed in the base of a cylindrical conductive cup of diameter 32 mm (Fig.5). Radon gas enters into the cup through a filter which prevents the inlet of radon progeny.*

*This device has the unique characteristics of a mobile wall which produces a syringe effect and an on-off switching of detector exposure. The pumping function allows fast radon transfer inside the cup and fast extraction at the end of the measurement interval. The new ENEA dosimeter offers many advantages: self protected storage, timed monitoring and easier calibration* *protocols. Contrary to conventional integrating passive alpha track detectors, the ENEA Piston Radon Dosimeter allows to start and stop exposure at well defined time intervals, e.g. during the stationary conditions of the reference atmosphere. To meet this aim prototypes have been constructed to be fitted on the three coupling flanges of the door of the exposure facility. This experimental set up allows to control the exposure interval of each detector from outside the test vessel (Fig.4).*

*Figure 6 shows the schematic of a radon exposure facility based on both real time monitoring of radon concentration and on the application of a set of calibrated integrating passive radon detectors which allows direct monitoring of exposure during each calibration run. Figure 7 shows the detailed schematic of the experimental apparatus with the lay-out of PRDs used to test the radon exposure uniformity inside the climatic chamber.*

*Experimental protocols for testing passive integrating detectors with radon reference atmospheres*

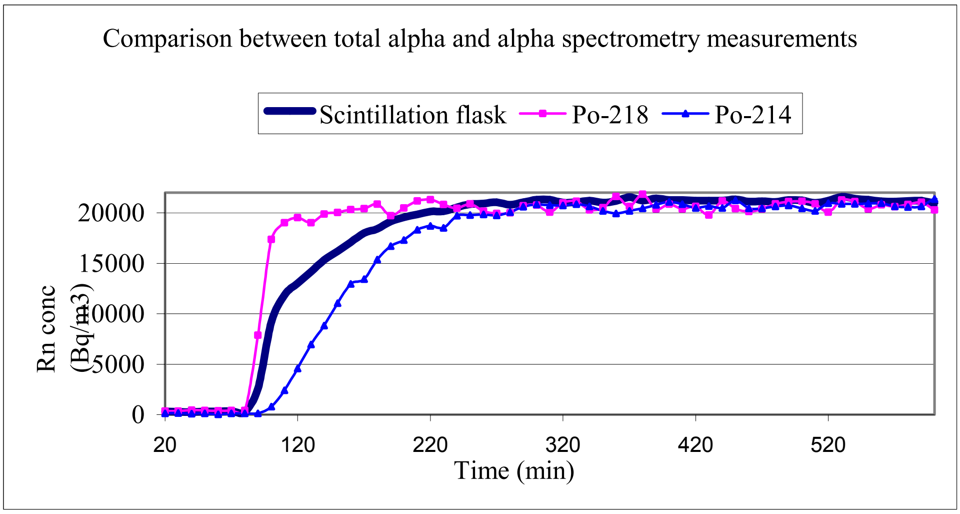


*Fig. 8 Build up of radon concentration inside the radon chamber at closed circuit conditions.*

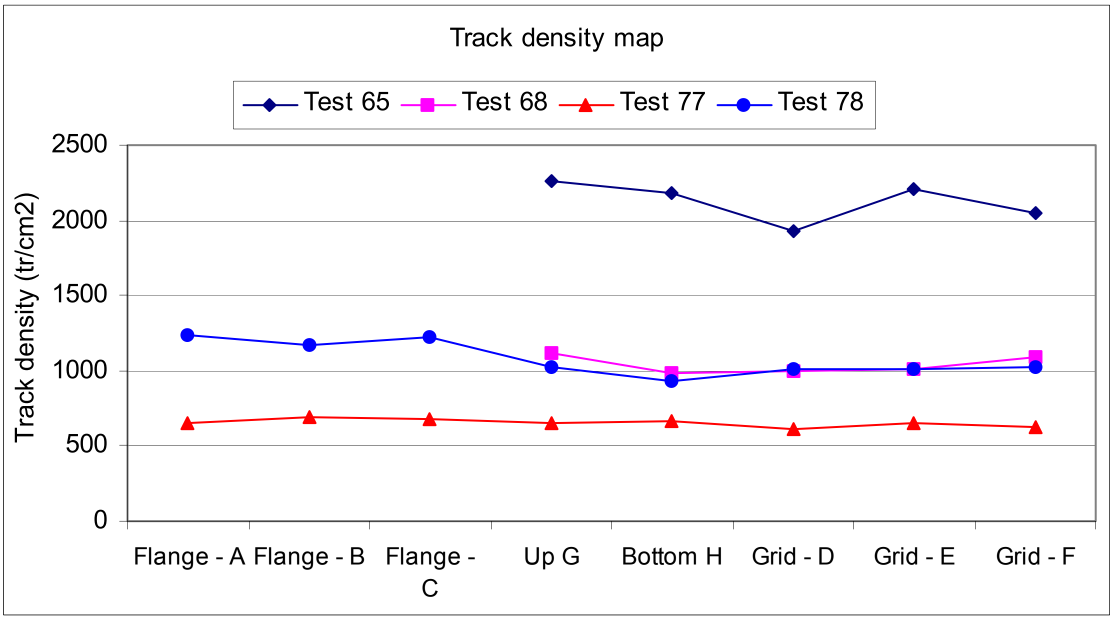

*Fig. 9. Track density distribution of PRDs on the flanges and inside the radon chamber for different experimental runs*

*Figure 8 shows the build up of radon concentration inside the radon chamber at closed circuit conditions, monitored with two different techniques, based on, respectively, total alpha counting of radon progeny decays and $^{218}Po$ and $^{214}Po$ spectrometric measurements. Experimental data show that a constant radon concentration is reached only after an initial transient. The calculation of radon exposure during the initial transient is less accurate than at stationary conditions (constancy of radon concentration). When calibrating ATDs, the reference atmosphere should be tested on two aspects: radon concentration uniformity and radon concentration stability during a calibration run. Figure 9 shows the track density pattern obtained positioning PRD sets at different points of the climatic radon chamber. The relative standard deviation of the mean based on the response of different PRD sets shows values inside the range: 1.5-4.5 %. The radon concentration within the exposure chamber resulted to be sufficiently uniform so that the time integrated concentration between the maximally and minimally exposed monitors differs by less than 5%.*

*Reproducibility tests of the PRD response to the same exposure level (1400 kBqh/m$^3$)*

*Based on four runs at open and one at closed circuit conditions have been carried out.*

*The response given in terms of average track densities per unit of exposure (calibration factor). The relative standard deviation for the above four experimental tests at open circuit conditions is 3%.*

*Tab1. Comparison between two different procedures used to evaluate radon exposure during a calibration run of passive alpha track detectors.*

| Reference exposure kBqh/m$^3$ | Uncertainty | Experimental exposure kBqh/m$^3$ | Uncertainty | Difference (percentage) |
|---|---|---|---|---|
| 1521 | ± 5,1% | 1560 | ± 13,0% | 2.6% |

*The table 1 shows the comparison between the exposure values obtained with two different procedures evaluated in the same time interval during the experimental run. The column 1 shows the exposure value obtained by the numerical integral of continuous measurements of radon concentration carried out with the scintillation cell.*

*The column 3 shows the exposure value obtained by the direct measurement with the passive time integrating technique, based on the use of alpha track detectors. Results evidence that measurements of radon exposure based on the two different techniques differ of about 2.6%.*

## *7. Final remarks*

*The approach of ENEA-INMRI on radon metrology has been based on a complete and integrated system which can be used to calibrate the main types of $^{222}Rn$ in air measuring instruments with international traceability. This* *system comprises primary and secondary standards for $^{226}$Ra and $^{222}$Rn activity, transfer standards, and reference instruments. The $^{222}$Rn Measurement System allows to calibrate the $^{222}$Rn Reference Instrument, used for the standardization of the radon test chambers with traceability to the national $^{226}$Rn primary standard. All the electronic equipment is based on the Radotron system which manages data-acquisition and control functions. Radotron is operated running a custom software, which automates evaluation and monitoring procedures. The climate test chamber has the ability to furnish a test atmosphere for radon monitoring equipment at controlled conditions. The unique characteristics of climate technology of the test chamber allow fully automated conditioning cycles, that doesn't influence the aerosol concentration and size distribution of reference atmosphere.*

*The PRD technique revealed to be a useful tool to check the radon concentration uniformity of the reference atmosphere. In the case of the ENEA exposure facility the radon concentration resulted to be sufficiently uniform so that the time integrated concentration between the maximally and minimally exposed monitors differs by less than 5%. The unique characteristics of the new ENEA PRD (i.e. fast sampling, negligible post-exposure effect and control of exposure interval) allows to optimize calibration procedures. A well calibrated PRD set on the door flanges of an exposure facility allows direct monitoring of exposure during each calibration run of passive integrating radon detectors. When used as a reference monitor of average radon concentration, the RPD set appears to be a very promising solution of a Radon Exposure Standard (RES) of good metrological quality.*

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