A laboratory facility (RTD) to study radon transport through modeled soil bed: results of preliminary measurements

Krzysztof Kozak, Jadwiga Mazur, Hayk Hovhannisyan

Abstract. The paper describes a laboratory facility (radon transport device – RTD) which makes it possible to study radon transport. The measuring position is a vertical cylindrical vessel with a height of 202 cm and a diameter of 24 cm. It can be filled with sand, gravel or other soil materials to be studied. The facility is providing radon gas from the source to the studied material and making it possible to measure radon concentration at different vertical distances from the source. The parameters of the medium (temperature, humidity) can be measured at the same time. The preliminary measurements using the RTD with sand as medium are presented.

Key words: radon • transport • facility • diffusion

K. Kozak[∞], J. Mazur, H. Hovhannisyan Laboratory of Radiometric Expertise, The Henryk Niewodniczański Institute of Nuclear Physics, Polish Academy of Sciences, 152 Radzikowskiego Str., 31-342 Kraków, Poland, Tel.: +48 12 662 83 32, Fax: +48 12 662 84 58, E-mail: Krzysztof.Kozak@ifj.edu.pl

Received: 5 November 2008 Accepted: 29 January 2009

Introduction

Radon (²²²Rn), with a half-life of 3.82 d, is a gaseous radioactive decay product of the radium isotope ²²⁶Ra. In nature three radon isotopes occur (²¹⁹Rn, ²²⁰Rn, ²²²Rn). These isotopes are gases and may be released from ground and rocks. The paper deals only with ²²²Rn. The other natural radon isotopes may be disregarded due to their minimal concentration of ²¹⁹Rn in the environment and due to the short decay time of ²²⁰Rn.

Measuring radon concentration and modeling its transport is very important in radiation protection as well as in seismic protection. In seismic protection radon is known as a precursor of earthquakes. Retrospective analysis of recorded data of radon concentration in the Armenian National Survey of Seismic Protection showed that the probability of seismic realization of short-term radon precursors is 45% in the case of earthquakes (with magnitudes M < 5.0) which took place in the territory of Armenia [4], and for strong regional earthquakes (with magnitudes $M \ge 6.0$) this probability rises up to 54%. Probability of seismic realization of radon precursor is proportional to the number of realized anomalies (i.e. those anomalies, after which seismic events took place) and inversely proportional to the total number of similar anomalies [9].

High indoor air radon concentrations have been considered to be hazardous for human health. The highest radioactivity values found – more than 2000 Bq/m³ – pose a risk even larger than that from cigarette smoking [8]. The annual mortality from the exposure to radon

in buildings represents 9% of all deaths owing to lung cancer and 2% of all cancer deaths in Europe [3].

The health risks can be minimized if we know how radon migrates through ground and how it is transported from ground to houses. Measuring radon concentration and modeling the radon transport give the possibility to solve the radon problem.

Radon and thoron are formed in the ground during radioactive decay of radium isotopes (²²⁶Ra and ²²⁴Ra). Radon concentration at any point of measurement is a function of the following factors [2]:

concentration and distribution of radium in the ground;

- physical and chemical properties of the ground;
- radon transfer from deep ground layers.

There are three stages from the formation moment of radon atoms to their escape from soil to the ambient air [5]:

- 1. Releasing of radon atoms from solid grains as gas or liquid in the pore spaces (emanation).
- 2. Transport of radon atoms through pore spaces (molecular diffusion, convection).
- 3. Escaping of radon atoms from soil to air (exhalation).

The fraction of radon atoms, generated in soil grains and reaching the pore volume of the soil, is known as

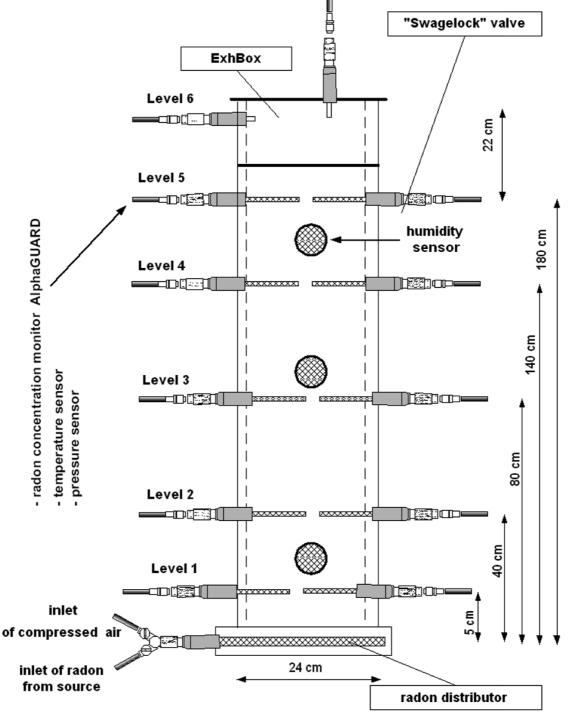


Fig. 1. Schematic view of RTD design.

the emanation coefficient. This coefficient depends on the soil grain size-distribution, radium content in soil, soil porosity and water content [6].

Diffusive transport is considered to be one of the main processes for exhalation of radon from the soil and is also considered to play a role in the transport of radon from building materials. Radon can be transported by convection, which is not limited to exchange of air between open spaces but it can also flow through porous media such as soil and building materials. As a result, porous media act as sources of convective radon. Convective transport is considered to be the most important mechanism for entry of radon from the soil to the living space [11].

The facility described in this paper gives the possibility to study diffusive and diffusive-convective radon transport as well as radon exhalation.

Equipment

The facility was designed and constructed by the authors with the technical assistance of Mr Tadeusz Zdziarski (Laboratory of Radiometric Expertise, Institute of Nuclear Physics PAN). The RTD has the form of a vertical cylindrical vessel of height 202 cm and diameter 24 cm. The vessel is made of PCV. It is filled with the required medium to the height of 183 cm. The top of the RTD is ended with the removable cover in which a Swagelock valve is mounted. This part of the cylinder (ExhBox) can be used for the measurement of radon exhalation from the medium.

The diagram of RTD construction is shown in Fig. 1 and the real view in Fig. 2.

Radon source (type RN-1025 by PYLON), based on radium ²²⁶Ra is connected to the gas distributor (the set of metal pipes with small holes) located at the bottom of the cylinder. From there the radon gas can diffuse up

sibility to obtain different concentrations of radon. The special openings with valves, made at differ-

through the medium. The radon source gives the pos-

ent distances from the bottom (level 1 to level 6 (see Fig. 1)), enable to measure radon concentration, humidity and temperature of the medium at the same time. Sensors for measuring these parameters are protected with tubes made of wire mesh located inside the cylinder (Fig. 1). The professional radon monitor AlphaGUARD PQ 2000PRO (Genitron[®]) was used for measurements of radon concentration in the RTD at different levels above the radon distributor. The humidity and temperature of the medium were measured using the field meteorological station type LB-741. ThetaProbe ML2× probes were applied for humidity measurements and the thermometer LB-710T served for temperature measurements [10]. RTD can be ventilated using compressed air. The description of the equipment is presented in Table 1.

Radon gas is provided to the radon distributor in RTD from radon sources using a system of pipes and valves mounted on the control board (Fig. 3).

The same control board is applied for a radon calibration chamber used in our laboratory for calibration of radon detectors. The required radon activity A is obtained by selecting the appropriate time of the source closing t_z (after its ventilation). This time is calculated for each case using the formula [7]:

(1)
$$t_{z} = \frac{\ln\left(\left|1 - \frac{A}{A_{z}}\right|\right)}{-\lambda_{Rn}}$$

where: A_z – source activity; λ_{Rn} – radon decay constant (7.55 × 10⁻³ h⁻¹).

Measurements and results

For preliminary measurements, the pure mediumgrained sand (grain porosity 38%, particle density 1.67 g/cm³) has been selected. The grain-size distribution is shown in Fig. 4.

For all measurements, the temperature and humidity of the sand was constant: ca. 21°C and 27%, respectively.

Radium (²²⁶Ra) concentration in the sand was measured by means of low background gamma spectrometry with a NaI(Tl) detector after reaching radioactive equilibrium (²²⁶Ra/²¹⁴Bi) in the sample. The calculations were made using the "three-window" method [1] and the obtained result is 5 ± 0.2 Bq/kg. The natural background concentration of radon ²²²Rn in the sand was also measured and it amounted to 56 Bq/m³.

The first series of measurements (consisting of 4 steps) was performed with the "forced" transport of radon which means pumping the radon gas from the source. During each step, the radon concentration was measured at a different level of RTD using AlphaGUARD (AG) – the details of the measurement are shown in Table 2. Each step started with pumping the radon from the source to the RTD for 2 h. Then, the source was closed and the AG monitor registered radon concentration for the next several hours. After each step, the RTD was ventilated

Fig. 2. Real view of RTD.

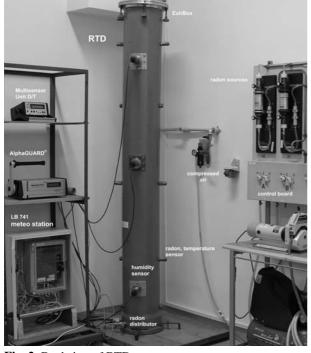


Table 1. Technical data of RTD equipment

PROFESSIONAL DEVICE FOR RADON MEASUREMENTS

AlphaGUARD PQ 2000 (Genitron[©])

Technical specifications

Radon concentration range: $2 \div 2,000,000 \text{ Bq/m}^3$ Range of temperature measurement: -10° C to 50° C Range of air humidity measurement: $0 \div 95\%$ rH Range of air pressure measurement: $700 \div 1100$ mbar Determination of the statistical error of radon concentration measurement Measurement and analysis of changeability tendencies in radon concentration

RADON FLOW THROUGH GAS SOURCES

RN-1025, nominal source activity 21.6 kBq, continuous radon output: 2.710 Bq/min RN-1025, nominal source activity 52.3 kBq, continuous radon output: 6.580 Bq/min

Technical specifications

Parent nuclide: rad (²²⁶Ra) Calibration accuracy: $\pm 4\%$ Flow rate (nominal): 0–10 litres/min Operating temperature: –20°C to +40°C Relative humidity: 0 to 100% Net weight: 1.7 kg Dimensions (length × width × height): 457 × 153 × 102 mm

MULTI SENSOR UNIT D/D (Genitron©)

Pressure difference (resolution) 0–500 Pa (2 Pa) 0–1250 Pa (5 Pa) 0–2500 Pa (10 Pa)

AIR-POWER PUMP (Cole-Palmer Instrument Co.)

Masterflex easy-load head Model 77601-10

FIELD METEOROLOGICAL STATION LB-741

Meteorological station LB-741 is designed for current control of the basic climate parameters (e.g. soil temperature and humidity).

The station includes a panel, which is attached with the following measurement devices: thermometer LB-710T, soil humidity meter ML2×, data registered in the station's panel can be read by the user using a PC computer through a communication interface of the station's panel (RS-232C – for local connection up to 20 m distance).

THETAPROBE SOIL MOISTURE SENSOR - ML2X [Th99]

Measured parameter: volumetric soil moisture content, $[m^3 \cdot m^{-3}]$ or [% vol.], device withstands burial in wide ranging soil types or water for long periods without malfunction or corrosion.

Technical specifications

Full range: from 0.0 to 1.0 m³·m⁻³ Accuracy: $\pm 0.01 [m^3 \cdot m^{-3}]$ for 0°C to 40°C, $\pm 0.02 [m^3 \cdot m^{-3}]$ for 40°C to 70°C Stabilization time: 1 to 5 s from power-up, depending on accuracy required Response time: less than 0.5 s to 99% of change Output signal: approx. 0–1 V DC for 0–0.5 m³·m⁻³ Case material: PVC Rod material: stainless steel

THERMOMETER LB-710T

Maximum scope of measurement: $-99.9^{\circ}C... + 199.9^{\circ}C$ Measurement inaccuracy for one-point calibration: $\pm 0.2^{\circ}C$ Measurement inaccuracy for two-point calibration: $\pm 0.1^{\circ}C$ Measurement resolution: $0.1^{\circ}C$

using compressed air. The results of this experiment are shown in Fig. 5. It can be seen that increasing radon concentration can be observed in all cases. However, the maximum radon concentration C_{max} and the time of reaching it depend on the distance from the radon distributor. In general, radon concentrations decrease with increasing distance from the bottom, mainly due to loss of kinetic energy and decay of radon atoms during diffusion through the medium. This is also the reason for decrease of the values of maximum radon concentration C_{max} with increasing distance from the radon source, therefore they can be reached after shorter times. The values of these parameters are given in Table 2. The lowest value of C_{max} was registered at the level 4 which is the most distant from the radon distributor. The slope of the curve of radon concentration

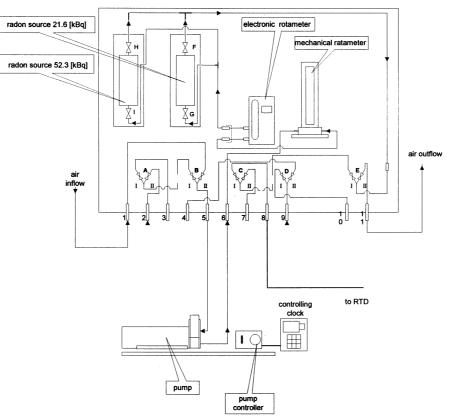


Fig. 3. Diagram of control board used for providing radon gas to RTD.

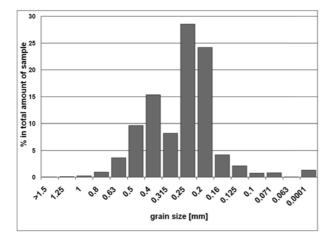


Fig. 4. Grain-size distribution of sand used in RTD.

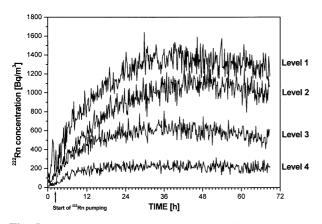


Fig. 5. The results of the first experiment ("forced" radon transport).

increase is also different for each level – the C_{max} is most quickly reached also at the level 4.

The second experiment was performed with the "non-forced" transport of radon. Three AlphaGUARD monitors were placed at different levels of RTD (see details in Table 3). The radon source was opened on 5th March 2008 and the radon gas could diffuse freely from the radon distributor into the volume of RTD. The source was closed on 11th March 2008. During all that time, AG monitors registered radon concentration at 10 min intervals. The results of this measurement are shown in Fig. 6.

The results show that the radon is transferred to the distances of 40, 80 and 140 cm during 8, 32 and 44 h respectively.

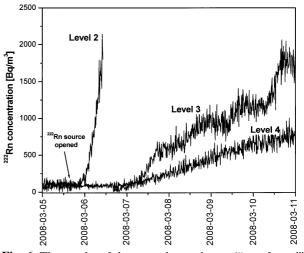


Fig. 6. The results of the second experiment ("non-forced" radon transport).

	Step 1	Step 2	Step 3	Step 4
Location of AlphaGUARD	Level 1	Level 2	Level 3	Level 4
	5 cm	40 cm	80 cm	140 cm
Rn pumping	28.01.2008	01.02.2008	08.02.2008	21.02.2008
from source start	12:15	16:10	11:20	17:10
(flow rate:	28.01.2008	01.02.2008	08.02.2008	21.02.2008
0.3 dm ³ /min) stop	14:15	18:10	13:20	19:10
Maximum radon concentration C_{max} (Bq/m ³)	1640 ± 120	1280 ± 120	810 ± 90	320 ± 50
Time of reaching C_{max} (h)	31	27.5	18.5	13

Table 2. Details of the first experiment ("forced" transport of radon)

 Table 3. Details of the second experiment ("non-forced" transport of radon)

Radon source	Open Closed	05.03.2008 16:00 11.03.2008 14:35
AG-1, Level 2 – 40 cm	start stop	04.03.2008 11:15 06.03.2008 10:30
AG-2, Level 3 – 80 cm	start stop	06.03.2008 15:40 12.03.2008 13:35
AG-3, Level 4 – 140 cm	start stop	04.03.2008 11:15 12.03.2008 13:35

Conclusions

The results of the preliminary measurements, using the new laboratory facility for investigation of radon transport RTD are presented in this paper. The design and construction of RTD as well as additional equipment are described. The test measurements showed that the presented setup may be applied for investigation of radon transport through sand which is a rather simple medium. The possibility of determination of diffusion constant for radon in particular medium was confirmed during the experiment with the "non-forced" radon transport.

The construction of RTD makes it possible to change media (e.g. different types of soil beds) and to investigate radon transport. Further measurements will be carried out for another types of media, and the properties of medium (e.g. humidity, temperature) will be changed during experiments. This will make it possible to study the dependence of radon transport on various parameters. The RTD will be also useful for the determination of contribution of diffusion and convection mechanisms in radon flux. One of the important advantages of the facility is also the possibility of investigation of radon exhalation from medium under controlled conditions regarding medium properties. The RTD is planned to be improved by applying pressure difference sensors and other types of radon source (e.g. uranium ore placed at the bottom of RTD instead of radon distributor).

Acknowledgment. The work was performed in the Laboratory of Radiometric Expertise IFJ PAN, Kraków, Poland.

References

- 1. Chiozzi P, Pasquale V, Verdoya M, De Felice P (2000) Practical applicability of field gamma-ray scintillation spectrometry in geophysical surveys. Appl Radiat Isot 53:215–220
- 2. Cothern RC, Smith JE Jr (1987) Environmental radon. Plenum Press, New York, pp 81–126
- Darby S, Hill D, Auvinen A et al. (2005) Radon in homes and risk of lung cancer: collaborative analysis of individual data from 13 European case-control studies. British Medical J 330(7485):223–227
- 4. Hovhannisyan H (2006) Probability of radon precursors of earthquakes (M < 5.0) in the territory of Armenia. In: Book of abstracts of the 1st European Conf on Earthquake Engineering and Seismology, 3–8 September 2006, Geneva, Switzerland, ID1626
- Janik M (2005) Radon transport model and its verification by measurements in houses. PhD thesis. Report IFJ PAN no 1966/AP. Institute of Nuclear Physics, Polish Academy of Sciences, Kraków (in Polish)
- 6. Jönsson G (1999) Experience from using plastic film in radon measurement. Radiat Meas 31:265–270
- Kozak K, Janik M, Mazur J et al. (2003) Radon calibration chambers at the Henryk Niewodniczański Institute of Nuclear Physics. Report IFJ PAN no 1920/AP. Institute of Nuclear Physics, Polish Academy of Sciences, Kraków (in Polish)
- Nazaroff WW, Nero AV Jr (1988) Radon and its decay products in indoor air. Wiley, New York
- Petrosyan H, Hovhannisyan H (2007) Probability of seismic realization of radon precursors of earthquakes (M ≥ 3.7) in Armenia and adjacent territories (abstract). In: International Union of Geodesy and Geophysics (IUGG) XXIV General Assembly, 2–13 July 2007, Perugia, Italy, SS005:6430
- ThetaProbe Soil Moisture Sensor type ML2×, User Manual (1999) ML2×-IU-1.21, Delta-T Devices Ltd
- Van der Pal M (2003) Radon transport in autoclaved aerated concrete. PhD thesis. Eindhoven University of Technology, Eindhoven, The Netherlands, University Press