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Intercomparison of radon CR-39 detector systems conducted in CLOR's calibration chamber

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Abstract. Six radon laboratories, which perform indoor radon measurements to jointly investigate temporal variations of radon concentration in houses in a couple of regions in Poland, participated in the intercomparison exercise. There are involved three commercially available Hungarian RadoSys systems and four own laboratory methods. All of them are based on the etched-track CR-39 detectors. The intercomparison was conducted in a calibration laboratory of the Central Laboratory for Radiological Protection (CLOR), Warsaw, Poland, accredited by the Polish Center for Accreditation (previously AP 101, at present AP 057). Comparison measurements were performed during three expositions in the CLOR's radon calibration chamber under controlled normal climatic conditions and steady radon concentration. The results were referred to the value of the radon concentration determined by AlphaGUARD monitor traceable to the primary ^{222}Rn standard in Physikalisch-Technische Bundesanstalt (PTB), Braunchweig, Germany. The mean performance ratio defined as a ratio of the reference to participant's result range from 0.88 to 1.31.

Key words: radon-222 • CR-39 track detector • intercomparison • RadoSys

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Introduction

Within the framework of the grant of the Polish Ministry of Science and Higher Education entitled Seasonal changes of indoor radon (^{222}Rn) concentrations of Poland (N506 1127 33) radon laboratories united in the Radon Center – the Non-governmental International Scientific Network – perform measurements of radon concentration in houses located in various areas of Poland. These laboratories belong to the following institutions:

1. Medical University of Białystok (MUB) in Białystok,
2. Wrocław University of Technology (WUT) in Wrocław,
3. Institute of Nuclear Physics, Polish Academy of Sciences (IFJ PAN) in Kraków,
4. Central Mining Institute (GIG) in Katowice,
5. Nofer's Institute of Occupational Medicine (NIOM) in Łódź,
6. Central Laboratory for Radiological Protection (CLOR) in Warsaw.

All laboratories use a passive time-integrating method of solid-state nuclear track detectors (SSNTDs) based on CR-39 foils, but they differ among each other with the origin of the detectors, design of diffusion chambers to house the detector, details in procedures of etching, track density counting systems and calibration factor. In order to get consistent results from all participants of the project an intercomparison exercise was arranged in the accredited calibration laboratory

AP 101 (at present AP 057) at the CLOR. Seven versions (from six laboratories) of the technique for radon concentration measurements, based on CR-39 detectors in diffusion chambers, participated in the intercomparison. The characteristics of the methods, devices and etching conditions are presented. The conduct of the intercomparison exercise is outlined below and is followed by a description of the calibration chamber and results.

About the etched-track techniques with CR-39 detectors

The etched-track technique based on a CR-39 foil is considered the most suitable method for indoor radon measurements. The foil CR-39, allyl diglycol carbonate, is a plastic polymer which has been used in the manufacture of eyeglass lenses since 1947. It was firstly applied during the World War II to coat aircraft fuel tanks for the B-17 bomber aircraft to make them more durable. The abbreviation stands for Columbia Resin #39 because it was the 39th formula of a thermosetting plastic developed by the Columbia Resins project (in 1940). During the early 1960s, the team of R. L. Fleisher, P. B. Price and R. M. Walker [4] revealed that CR-39 plastic is one of the SSNTDs in which heavily ionizing particles (e.g. alpha particles) penetrating the material cause damage to the chemical bonds along the track of the particle.

Upon etching process in a caustic solution of sodium hydroxide, the damaged regions become enlarged and visible under a conventional optical microscope as a distinguishable tracks which are counted by computerized systems. The track density is proportional to the time integrated exposure of the detector to alpha-emitting nuclide. The CR-39 material is the most sensitive plastic, if compared with others, to alpha particles and it records them in the widest energy range from ca. 0.1 to > 20 MeV [1]. It is not sensitive to beta and gamma radiation and correction for temperature and relative humidity conditions in the environmental range is not necessary. Unfortunately, atmospheric pressure may influence the track density because of a dependence of the alpha particle range on the air density. It was studied for another kind of alpha-track detectors Kodak LR 115 II published by L. Vasudevan and M. McLain [7]. They observed an inverse relationship of track density with air pressure (direct with altitude): for an altitude of 1500 m above the sea level the track density increased by 41%.

It is important to remember that the differences in production procedures of the CR-39 foils make significant differences in response to alpha radiation, that is to say in the calibration factor, for making radon measurements [3].

A convenient feature of CR-39 detectors from the point of view of their users is that, as it seems, they do not show "fading" of latent tracks in the time period between irradiation and etching [2]. Thus, detectors can be stored after exposition for a long time. An important advantage of all passive track-etched methods is that they give averaged overexposure period results of radon concentration.

For measurements of radon concentration in dwelling air, slices of the CR-39 foil are usually placed at the bottom of diffusion chambers. Such a chamber is made of an either conductive or non-conductive plastic cup. In the conductive plastic carbon is impregnated and this allows to avoid the problem of electrostatic charge. Areas of persistent charge inside the cup may be a source of artifacts that interfere with the accumulation of alpha tracks [5]. Some cups, e.g. of Kernforschungszentrum Karlsruhe (KfK) type, are covered with a fiber-glass filter to prevent radon decay products from entering and also limits thoron entry to the cup. Another kind of chamber, very often used, of English/Swedish (NRPB/SSI) type, later modified by RadoSys Kft., applies an air-gap filter. The chamber consists of only two plastic parts: a pot and a lid, which are a matching pair but not perfect. Radon gas, whose atom size is of the order of 10^{-4} μm can easily diffuse inside through the air-gaps of 10 μm. Radon progeny attach easily to aerosols and in their path, which is much longer than the width, get deposited in the wall. In consequence, the air-gap filter is transparent for the radon gas and constitutes a barrier for radon daughters which occur outside the chamber. Concentration of radon inside all types of the diffusion chamber is the same as outside and the CR-39 foil at the bottom of the chamber is struck by alpha particles emitted by radon and radon decay products which came into being from the radon inside the chamber.

It is worth mentioning that Swedish researchers [6] discovered that the plastic chambers absorbs radon which diffuses out of the plastic when chambers are stored in a lower radon concentration. If the CR-39 foils are not separated from chambers after exposure and stored in a closed environment the radon released during storage produces additional tracks in the foils what may generate overexposure error of measurement. The authors recommend that the films be separated from the chambers as soon as possible after the intended exposure.

Table 1. Characteristics of diffusion chambers and conditions of etching

Participant	Diffusion chambers				Etching conditions		
	Design	Conductivity	Kind of filter	Provider of CR-39	Concentration of NaOH (%)	Temperature (°C)	Duration (h)
MUB	RadoSys	Yes	Air-gap	RadoSys	20	92	4.5
GIG	RadoSys	Yes	Air-gap	RadoSys	25	90	4.5
IFJ PAN	RadoSys	Yes	Air-gap	RadoSys	25	90	4.5
CLOR1	KfK	No	Fiber-glass	TASL	40	70	7
CLOR2	SSI	Yes	Air-gap	TASL	40	70	7
NIOM	NRPB	No	Air-gap	Pershore Lmt.	22	80	18
WUT	TASL	Yes	Air-gap	TASL	25	75	6

Table 2. Characteristics of methods

Participant	System of counting	Counted area (mm^2)	Bkg (tracks/ $mm)^2$	CF $\left(\frac{kBq \cdot h \cdot m^{-3}}{tracks/mm^2}\right)$	Uncert. (%)	LLD ($kBq \cdot h \cdot m^{-3}$)	Uncert. (%)	Min. measurable Rn concentration ($Bq \cdot m^{-3}$)	
								for 30 day exp.	for 90 day exp.
MUB	RadoSys	46	0.36	30.3	3	15	12	21	7
GIG	RadoSys	46	0.3	43.35	3	25	60	26	9
IFJ PAN	RadoSys	46	0.3	36.01	3	16	12	23	8
CLOR KfK	Own	78	1.5	31.2	8	50	20	69	23
CLOR SSI	Own	78	1.5	50.7	8	80	20	111	37
NIOM	Own	44.1	1.5	40	2.9	20	45	28	9
WUT	Own	98	0.2	155	12	35	35	49	16

Seven versions of the track-etched method participated in the intercomparison. In Table1 there are given data on the used devices: their design, conductivity, kinds of filter and also conditions of etching: concentration of sodium hydroxide, temperature and duration. Table 2 presents the characteristics of each method such as: counted area, background density, calibration factor (CF) and low level of detection (LLD).

Laboratory facilities

The intercomparison exercise was conducted in the CLOR calibration facilities. It is a walk-in climatic chamber (Fig. 1) of inner volume of 12.38 m^3. The chamber has an anteroom of 3.12 m^3, a viewing heated window, manipulating gloves and a number of input ports which permit injection of radon gas and aerosols, sampling inside air and connecting instruments (e.g. aerosol counter) outside. The chamber is equipped with a ventilation system to remove the radon outdoors after experiments. Climatic conditions, temperature and relative humidity (RH) can be steered automatically. Temperature may be set up from –30°C to +60°C at constant time ± 1°C and RH from 10 to 95% with a long time margin of ± 5% (for the temperature range from +10°C to +60°C).

To create radon atmosphere in the chamber, one of two external radon generators, the dry flow-through ^{226}Ra sources of 137.27 kBq or 502.5 kBq manufactured by Pylon Electronic Development Inc., Canada, is connected to two ports on the opposite sides of the chamber.

Fig. 1. The walk-in radon calibration chamber of CLOR.

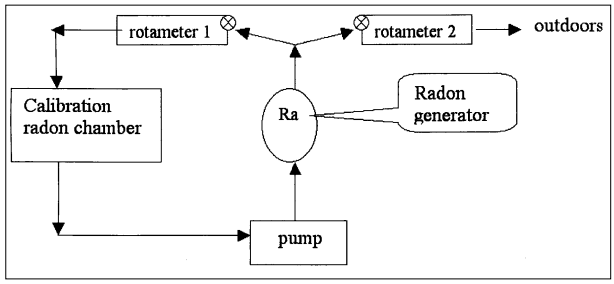

Fig. 2. Scheme of the fork type connection system of the radon generator with the calibration chamber.

There is a possibility of work in two modes: mode of one-time supply of radon produced up in the generator and the continuous supplying mode. In the first mode the concentration of radon in the chamber decreases during an exposure with the half-life of the radon decay. In the second mode the radon concentration is kept steady by means of continuous pumping. The following steady concentrations are achievable: maximum concentrations corresponding to the activity of radium sources (ca. 10, 40 and 50 kBq/m^3 for a series connection of two) and any lower concentrations which can be achieved with the use of a fork type connection system: tree-way connector and two rotameters with adjustment of flow rates placed at two sides of the connector – one on the way to the chamber and another one on the way outdoors. The scheme of the fork type connection is presented in Fig. 2. By adjustment of the two flow rates in relation to each other, it is possible to create almost steady radon concentration in the chamber at the level of the initial concentration at a required level. An example of the time course of the steady radon concentration for the initial concentration of ca. 3700 Bq/m^3 is shown in Fig. 3.

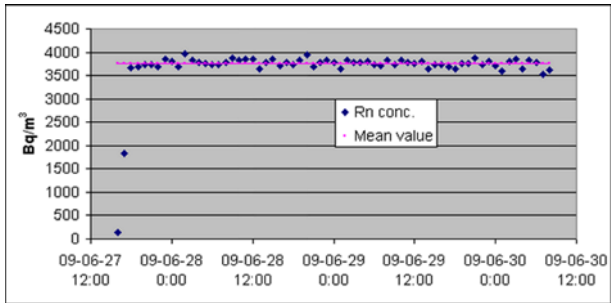

Fig. 3. The time course of radon concentration in the radon chamber with use of stable pumping in the fork type connection system.

Fig. 4. Exposition of diffusion chambers with CR-39 being compared in the radon chamber.

The conduct of the intercomparison experiment

It was planned to conduct three exposures. Participants delivered 20 detectors for each method: five detectors for each exposure and five as transport background detectors. All the detectors to be compared were placed in the calibration chamber (Fig. 4). The first batch was taken out of the chamber after 41 h, the second one – after 93 h and the third – after 161 h. During the exercise, a radon generator of 137.27 kBq was applied and two flow rates were adjusted to each other to keep the radon concentration in the chamber within limits of 5000–4600 Bq/m^3. Hourly averaged radon concentrations were registered by means of the reference device – AlphaGUARD PQ 2000 PRO monitor manufactured by Genitron Instruments GmbH, Germany, with the calibration coefficient traceable to PTB. In Table 3 there are presented: reference values of three exposures: *A*, *B* and *C*, mean values of radon concentration and climatic parameters corresponding to each exposure according to AlphaGUARD PQ 2000 PRO readings.

Results and discussion

Table 4 gives the results delivered by the participants, performance ratios defined as the reference value divided by the participant's result with an associated uncertainty calculated from the uncertainty of both values and E_n-test values calculated from the following formula:

$$E_n = \frac{R_{\text{ref}} - R_{\text{part}}}{\sqrt{u_{\text{ref}}^2 + u_{\text{part}}^2}} \quad (1)$$

where: E_n – a coefficient for estimation of consistency of results; R_{ref} – reference value; R_{part} – result of a participant; u_{ref} – uncertainty of the reference value; u_{part} – uncertainty of the participant value.

The performance ratio ranges from 0.91±0.09 to 1.12±0.26 in the exposure *A*, from 0.83±0.13 to 1.38±0.48 in the exposure *B* and from 0.85±0.11 to 1.15±0.20 in the exposure *C*.

Interpretations of E_n-test values recommended by the Polish Center for Accreditation (PCA) assumes that the results are consistent if $|E_n| \leq 1.00$.

It is interesting to compare the two methods of estimations of the spread of results. For only one result (VII in the exposure *B*), the $|E_n|$ value equal to 1.08 exceeds the number 1.00, which means, it does not fulfill the criterion of PCA. The performance ratio for this case is 1.38±0.48. For performance ratios of 0.83, 0.85 or 1.15, which seem to be not very satisfactory, the E_n-test values are equal to 0.6, 0.7 and 0.9, respectively, which means these results satisfy the PCA criterion for results to be consistent. This is due to a strong dependence of E_n-test value on uncertainty of the results. E_n-test value may satisfy the criterion even for a big spread of results, if uncertainties are high enough.

The results of the intercomparison are also illustrated graphically in Fig. 5.

Table 3. Values of exposures, duration, means of radon concentration and climatic parameters during intercomparison expositions

Symbol of exposure	Duration (h)	Mean Rn concentration (Bq·m^{-3})	Exposure (kBq·h·m^{-3})	Pressure (mbar)	Temperature (°C)	RH (%)
A	41	4974 ± 317	204 ± 13	998	25	45
B	93	4619 ± 290	430 ± 27	1003	25	54
C	161	4594 ± 286	740 ± 46	1007	25	58

Table 4. Results of participants, performance ratios (PR) and E_n-test

Participant	Experiment *A*			Experiment *B*			Experiment *C*			Mean of PR
	Results (kBq·h·m^{-3})	PR	E_n-test results	Results (kBq·h·m^{-3})	PR	E_n-test results	Results (kBq·h·m^{-3})	PR	E_n-test results	
I	217 ± 12	0.94 ± 0.08	0.77	424 ± 28	1.01 ± 0.09	0.15	710 ± 23	1.04 ± 0.07	0.58	1.00
II	205 ± 17	1.00 ± 0.10	0.03	440 ± 30	0.98 ± 0.09	0.25	705 ± 43	1.05 ± 0.09	0.56	1.01
III	224 ± 16	0.91 ± 0.09	0.97	472 ± 33	0.91 ± 0.09	0.99	793 ± 47	0.93 ± 0.08	0.81	0.92
IV	213 ± 32	0.96 ± 0.15	0.27	439 ± 64	0.98 ± 0.16	0.13	740 ± 106	1.00 ± 0.16	0.00	0.98
V	215 ± 32	0.95 ± 0.15	0.33	422 ± 60	1.02 ± 0.16	0.13	701 ± 101	1.06 ± 0.17	0.35	1.01
VI	210 ± 76	0.97 ± 0.36	0.08	520 ± 150	0.83 ± 0.24	0.59	870 ± 190	0.85 ± 0.19	0.66	0.88
VII	172 ± 36	1.12 ± 0.26	0.51	287 ± 44	1.38 ± 0.48	1.08	593 ± 82	1.15 ± 0.20	0.89	1.31

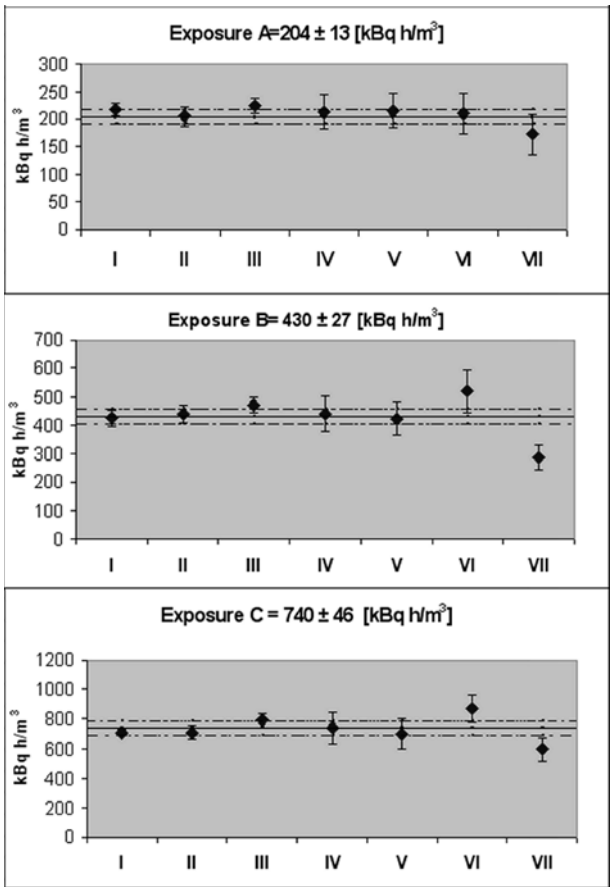

Fig. 5. Results of the intercomparison of seven versions of the track-etched method based on CR-39-detectors.

Summary

For five of twenty one results the performance ratio exceeds 10% of unity. For one laboratory the mean performance ratio exceeds 30% and the E_n-test is not satisfied in one of three experiments. Thus, it can be said that the results of the intercomparison of the CR-39 detector methods applied in seven radon laboratories are in good order.

The authors agreed that if the mean value of performance ratios for three exposures is in the range from 0.90 to 1.10, participants do not need to correct their calibration factors. If it goes out of the range they should correct it.

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References

1. Cartwright BG, Shirk EK, Price PB (1978) A nuclear track recording polymer of unique sensitivity and resolution. Nucl Instrum Methods 153:457–460
2. Durrani SA, Bull RK (1987) Solid state nuclear track detection – principles, methods and applications. Pergamon Press, Oxford, p 129
3. Espinosa G, Dudney CS, Gammage RB (1995) Intercomparison of response characteristics of seven CR-39 materials during radon calibration. Radiat Meas 25;1/4:611–612
4. Fleisher RL, Price PB, Walker RM (1965) Solid-state track detectors: applications to nuclear science and geophysics. Ann Rev Nucl Sci 15:1–28
5. George AC (1996) State-of-the-art instruments for measuring radon/thoron and their progeny in dwellings – a review. Health Phys 70;4:451–463
6. Möre H, Hubbard M (1997) ^{222}Rn absorption in plastic holders for alpha track detectors: a source of error. Radiat Prot Dosim 74:85–91
7. Vasudevan L, McLain M (1994) Atmospheric pressure effects on the calibration constant of alpha-track radon detectors. Health Phys 66;3:318–326