RETROSPECTIVE RADON MEASUREMENTS BASED ON IMPLANTED ^{210}Po IN GLASS OBJECTS USING POLYCARBONATE DETECTORS

by

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Scientific paper
http://doi.org/10.2298/NTRP1802167A

In the present investigation, a surface-deposited polonium was measured in 37 houses in Rasht and Ramsar cities of Iran with the aim of evaluating the retrospective radon concentration. The CR-LR technique is widely used in this regard, but for the first time, Lexan polycarbonate detectors were used to measure the activity of ^{210}Po planted in glassy objects. These detectors were placed on glassy surfaces for 153 to 365 days. A passive cylindrical diffusion chamber was used for the contemporary radon concentration measurements. The diffusion chamber consists of the Lexan polycarbonate films as a solid state nuclear track detectors and filter. The surface-deposited ^{210}Po activity concentration was found to vary from 0.26 to 11.96 mBqcm^{-2} with average of 2.62 mBqcm^{-2}. The sensitivity of ^{210}Po to polycarbonate was determined to be 0.06456 track per cm^2 per mBqhcm^{-2}. Thus, the radon concentration was found to vary from 122 to 4840 Bqm^{-3} with an average value 1243 Bqm^{-3} and the contemporary radon concentration in the area was found to vary from 15 to 2420 Bqm^{-3} with an average 513 Bqm^{-3}. The results indicate that there is a significant correlation between the concentration of the retrospective radon and the concentration of the contemporary radon gas in the indicated areas with a coefficient of 0.80672.

Key words: glassy object, retrospective measurement, polonium 210, polycarbonate detector, radon

INTRODUCTION

The radon gas is one of the most important sources of natural radiation and a high exposure to it can lead to the lung cancer [1, 2]. The methods for long-term passive radon measurements based on the nuclear track detectors have been very well established and widely used. The surveys of existing techniques have been given by Nikolaev and Ilic [3] and Nikezic and Yu [4]. The long-term measurements were held for various periods of time, ranging from 3 to 12 months [5]. The measurements of contemporary radon in homes to assess the radiation levels of individuals in the past are not enough because this time is still too short compared to the average lifespan of a person [6, 7]. However, the factors such as seasonal changes can have an impact; also, due to the architectural changes in the houses or the change in owner's behavior, such as ventilation rates or the use and the non-use of cigarettes, the concentration of indoor radon can be different [5, 8]. Thus, measurements performed for a few months might not be a suitable representation of "real" indoor radon concentration. As a possible solution to this problem, the retrospective dosimetry based on the measurements of ^{210}Po activity in objects was proposed. The retrospective radon dosimetry for home environments based on the measurements of implanted ^{210}Po activities in glass objects have been introduced and practiced for many years [9]. Nikezic and Yu described the behavior of the short-lived radon progeny on surfaces of objects, leading to their deposition and implantation after alpha-particle decays. The radon decay chain includes the short-lived radon progeny ^{218}Po, ^{214}Pb, ^{214}Bi, and ^{214}Po. Their successor, ^{210}Pb, which is a long-lived radon progeny with a half-life of 22.3 years, can be implanted into the glass surfaces. Thus, the activity increases with the time of exposure of the object to short-lived radon progeny. The second successor of ^{210}Pb is the alpha emitting nuclide ^{210}Po, whose activity in the subsurface layer can be determined by the alpha-particle measurements [10]. The methods were

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developed for determining the retrospective ^{222}Rn concentration based on the measurements of ^{210}Po activity in an object by using various techniques of α-activity measurements [11-15]. A retrospective study that was developed in recent years used the surface trap method. In this technique, the α-recoil implanted ^{210}Po on the glass and other vitreous artefacts were measured using the solid state nuclear track detectors [16, 17]. The activity of implanted ^{210}Po in the glass is related to the concentration of ^{222}Rn gas [10].

The present study was conducted in Rasht and Ramsar cities of Iran with the aim of evaluating the retrospective radon gas concentration based on the activity of ^{210}Po planted on glass objects in 37 houses, and selected glass objects were obtained from ages 8 to 50 years. Ramsar was selected because it is a high natural radiation area. Ramsar, a northern coastal city in Iran, has areas with the highest levels of natural radiation measured to date. The effective dose in the high level natural radiation areas (HLNRA) of Ramsar, in particular, Talesh-Mahalleh district, are few times higher than the International Commission on Radiological Protection (ICRP)- recommended radiation dose limits for radiation workers is. The annual radiation effective dose from the background radiation in HLNRA of Ramsar is up to 260 mSv. The high background radiation in the "hot" areas of Ramsar is primarily due to the presence of very high amounts of ^{226}Ra and its decay products, which are brought to the Earth's surface by hot springs. It is worth mentioning that the radon levels in some regions of Ramsar are up to 3700 Bqm^{-3} [18-21].

Also, a comparison was made between the concentration of the contemporary radon gas and the retrospective radon gas concentration.

MATERIALS AND METHODS

In this research, a glass-based measurement with the surface trap retrospective method was used. The 250 m thick (2.5 cm × 2.5 cm) Lexan polycarbonate, as Solid State Nuclear Track Detector (SSNTD) was used in this study to register alpha particles for the first time. The surface trap method was used because the process does not damage objects inside the house like the volume trap method, and the Solid State Nuclear Track Detectors were used because they are easily found and due to the cheapness, the phenomenon of fading does not occur and it is not brittle, and the effects of radiation on them are permanent [22].

The polycarbonate detector was placed on the glass surface to record the alpha particles emitted from the ^{210}Po. The ^{210}Po has an alpha energy of 5.3 MeV, while the efficient registration of alphas in PC are in a range about 0.5 to 2 MeV using the electrochemical etching (ECE) for PEW etchants [23, 24]. The PEW solution including the KOH (15 %), C_2H_5OH (40 %) and H_2O (45 %) and the ECE process of 32 kVcm^{-1}, 2 kHz and 3 h was applied as an effective method of alpha particles detection in PC [25]. For this reason, the number of degrader layers was calculated using the Trim software and based on the results of the experimental measurement, four degrader layers made of aluminum foil (Mylar) were attached on the PC foils. After preparing the detector and cutting it into 2.5 cm × 2.5 cm, it was installed on the selected glass surfaces. As shown in fig. 1, B is a Lexan polycarbonate detector and A is a 4-layer aluminum foil. In the selection of glass surfaces, the glass lifetime of 10 years was considered and also the glass surface was not in the wind stream. In addition, the selected glasses were flat and not exposed to the sunlight. According to the previous studies [13, 16, 26, 27] only some types of glass objects were considered suitable, such as mirrors, picture frames, paintings or large photos, cabinet glasses, glasses in internal doors, and glasses in large wall clocks. Moreover, other materials, such as the ceramic objects were not considered suitable because of the uncertainties in the background activity in the surface glazes [28].

In this study, a total of 18 glass objects in Rasht and 19 glass objects in Ramsar were selected in residential houses of these two cities. The detectors made according to fig. 1 were installed on the glass surface of the selected objects. After a specified period of time, all the installed detectors were collected. The electrochemical etching was used to enlarge the latent tracks to the visible tracks. For this purpose, the detectors were placed between the two cells of the chamber. Then, the 3 % hydrochloric acid was added to one side of the chamber and the etchant PEW solution was introduced onto the other side of the chamber. After the electrochemical etching by applying a strength field of 32 kVcm^{-1} at 2 kHz frequency for 3 h, the detectors were taken out and the tracks density were measured [25, 29].

After counting the tracks, the activity of ^{210}Po implanted in glass was calculated using the following eq

$$A_{\mathrm{Po210}} \quad \frac{N_T \quad N_{T0}}{TK} \tag{1}$$

where, A_{Po210} is the ^{210}Po activity in terms of mBqcm^{-2}; N_{T} – the net number of tracks per cm^2 on the polycarbonate detector, N_{T0} – the background number

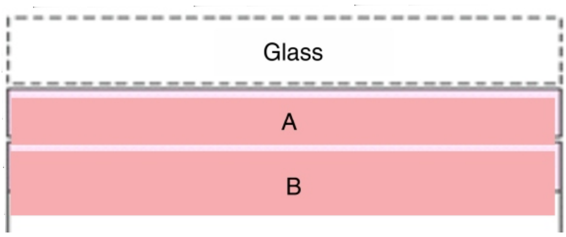

Figure 1. The installation method of a retrospective detector on the glass surface, where B is a Lexan polycarbonate detector and A is a 4-layer aluminum foil

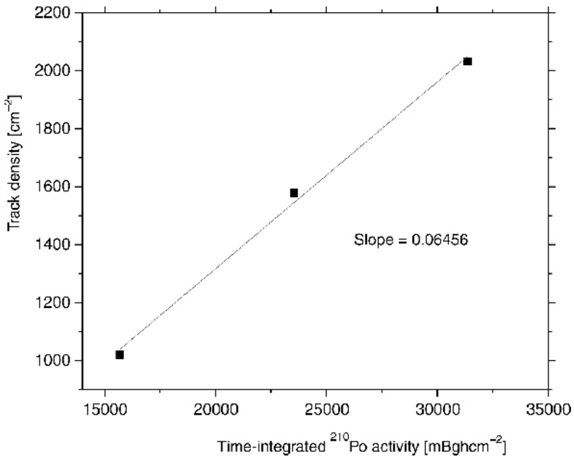

Figure 2. The linear relationship between the track density and the time integrated ^{210}Po activity (the slopes of linear curves were used to obtain the calibration factor (sensitivity))

of tracks per cm^2, T – the period of time [h] for which the detectors were mounted on the glass surface, and K – the sensitivity factor of polycarbonate detector to ^{210}Po activity which was obtained in calibration exercise and depends on the etching conditions [5, 28]. Figure 2 shows that there is a linear relationship between the track density and the time integrated ^{210}Po surface activity. The slope of this curve is 0.06456 track per cm^2 per mBqhcm^{-2}, representing the detector sensitivity. The polycarbonate films with 4-layer aluminum were placed on the surface of ^{241}Am standard disc source. The time of exposure was 2, 3 and 4 hours. Since the energy of alpha of ^{241}Am (5.4 MeV) is close to the ^{210}Po (5.3 MeV), the ^{241}Am standard source was used for calibration in this study.

The surface activity of ^{241}Am standard source was 7.8 Bqcm^{-2}. A disc source with the diameter of 5 cm was used.

Based on the activity of implanted ^{210}Po and the age of the glass objects, the retrospective radon concentration was estimated using the following expression [30]

$$C_{\text{Rn}} \quad 245\frac{A_{\text{Po210}}}{(1 \quad \text{e}^{\lambda_{\text{Po210}}t})} \quad _i \quad \frac{fi(P_i)}{fi(\overline{P_i})} \tag{2}$$

where C_{Rn} is the retrospective radon concentration in term of Bqm^{-3}, A_{Po210} – the ^{210}Po activity [mBqcm^{-2}], λ^{210}Pb – the decay constant for ^{210}Pb, with its amount being 0.031 per year, t – the age of the glass object [year], P_i and $\overline{P_i}$ are real values and the values of global average related to room parameters, that is, the concentration of aerosol particles, the deposition velocity of the unattached ^{218}Po, ^{214}Po, ^{214}Pb and the deposition velocity of attached ^{218}Po, ^{214}Po, ^{214}Pb, the air exchange rate and the surface to the volume room ratio. Fi is the fitted function related to each parameter that has been provided by Bastrikov, *et al.* (2006)[30]. This equation is correct for the exposure duration t T $_{\text{Po}}$

Of course, in this study, considering the limitation of measurement devices, the uncertainty and the most influencing model parameters, the fitted functions related to the concentration of aerosol particles (Z, cm^{-3}) and the air exchange rate (λ_{V}, h^{-1}) and the surface to the volume room ratio (S/V, m^{-1}) for measurement were the only ones considered and they are as follow

$$f_1(Z) \quad 2.47.Z^{0.331} \tag{3}$$

$$f_2(\lambda_{\text{V}}) \quad 47.1 \quad 7.58.\lambda_{\text{V}}^{0.686} \tag{4}$$

$$f_3 \quad \frac{s}{V} \quad 38.6 \quad 2.64\frac{s}{V} \tag{5}$$

These models are sensitive to the values of a number of key room parameters. These are principally the characteristics of the room aerosols, the room surface to the volume ratio and the ventilation rate. In a large-scale survey of dwellings, the range of these parameters in the past was not known and even their present values can only be estimated. However, there is still an uncertainty in this approach [30, 31].

RESULTS AND DISCUSSION

Out of the 37 detectors mounted on the glass surfaces in 37 houses, 18 belonged to Rasht, being named with codes a_1 to a_{18}, and 19 belonged to Ramsar, being named codes b_1 to b_{19}. One of the main requirements in low concentrations to provide the reliable measurements is determination of the Minimum Detection Limits. The minimum detection limit (MDL) was obtained using eq. (6) [32]

$$\text{MDL}_{(\text{Po210})} \quad \frac{2\sigma}{(k \quad T)} \tag{6}$$

where σ is the standard deviation of the net track density of the unexposed Lexan film, k – the calibration factor, of the measurements (the track density per exposure) and T – the time of exposure (days). According to the eq. (6), the MDL (^{210}Po) for 7 months was calculated about 0.2290 mBqcm^{-2} or 1146 mBqcm^{-2}h^{-1}. So, the codes a_1, a_2, a_3, a_4, a_6, a_7, a_9, a_{11}, a_{13}, a_{14}, and b_9 with values below the above MDL were deleted. Therefore, out of the 37 data, 26 were confirmed. Thus, the actual sample included 26 dwellings, and 26 suitable objects were considered (4 mirrors, 13 picture glasses, 5 cabinet glasses, 3 glasses in interior doors, and 1 glass in large wall clocks). The average age of the glass objects was 23 years. The results of this study are presented in tab. 1. The concentration of ^{210}Po in household glass objects was found to vary from 0.26 to 11.96 mBqcm^{-2} with an average of 2.62 mBqcm^{-2}.

For each of the 26 glass objects, the concentration of the retrospective radon based on the activity of the implanted ^{210}Po in glass calculated using the Equation

Table 1. The activity of ^{210}Po for residential buildings in Rasht and Ramsar based on the age of the glass

Number	Code	Type glass	Age glass (year)	Day	A_{Po210} [mBqcm^{-2}]
1	b_1	Picture glass	8	182	2.1702
2	b_2	Picture glass	10	167	1.3197
3	b_3	Picture glass	10	177	1.9288
4	a_5	Mirror	10	365	0.3562
5	b_4	Mirror	12	156	0.5647
6	a_8	Mirror	12	352	0.2594
7	b_5	Picture glass	15	176	0.4620
8	b_6	Picture glass	15	174	0.8976
9	b_7	Cabinet glass	15	176	2.7960
10	a_{12}	Cabinet glass	18	295	0.7755
11	b_8	Picture glass	19	159	2.3015
12	b_{10}	Mirror	20	164	0.6670
13	a_{10}	Cabinet glass	22	365	0.8407
14	b_{11}	Picture glass	25	172	1.6566
15	b_{12}	Picture glass	25	177	1.6754
16	b_{13}	Picture glass	26	182	0.9964
17	b_{14}	Glass of clock	29	182	1.9982
18	b_{15}	Picture glass	30	183	10.53
19	b_{16}	Door glass	30	155	10.4
20	b_{17}	Door glass	30	155	11.96
21	a_{15}	Cabinet glass	31	365	0.3898
22	a_{16}	Picture glass	35	295	0.6508
23	b_{18}	Picture glass	35	153	5.1041
24	a_{17}	Picture glass	40	352	0.3566
25	a_{18}	Cabinet glass	42	295	0.6147
26	b_{19}	Door glass	50	157	6.4820

(1) was between 122 to 4840 Bqm^{-3} with the mean of 1243 Bqm^{-3}, while the concentration of the contemporary radon gas in these regions was 15 to 2420 Bqm^{-3} with the mean of 513 Bqm^{-3}. The comparison of the retrospective and the contemporary radon concentration values for each of the codes is shown in fig. 3.

As shown in fig. 3, there is a significant difference between the concentration of the retrospective radon and the concentration of the contemporary radon gas inside these houses. In all the cases, the retrospective radon concentration was higher than the contemporary radon concentration. The average retrospective radon concentration obtained in 26 houses was 1243 Bqm^{-3} and the contemporary radon concentration was 513 Bqm^{-3}. Thus, the average retrospective radon concentration is 2.4 times more than the average contemporary radon concentration, which can be attributed to the change in house conditions over the period of 23 years. These changes may be due to the change in the architecture of the home or the change resulting from the renovation of the houses or the changes in the household appliances, such as changing the heating system from gas heaters to radiators or changes in the ordinary individual's life, such as changing the ventilation rate, or even changing the alpha particle planting rate on the glass surface. However, the location of glass objects within the houses also influences the retrospective radon concentration because, sometimes, the object is implanted in the location where the planting activity is found more.

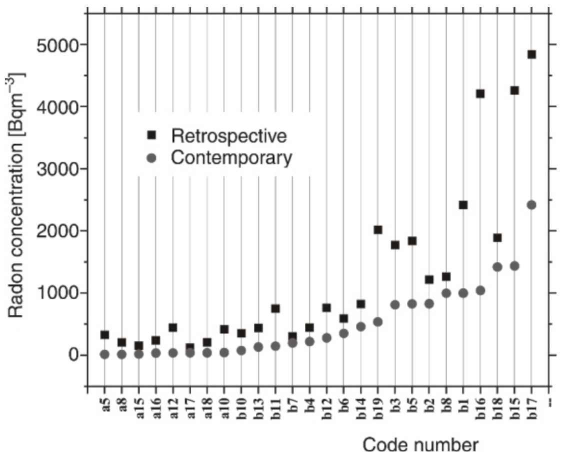

Figure 3. The retrospective and the contemporary radon concentration chart based on the house codes

Of course, changes in room parameters, such as the concentration and the size of distributed aerosols, the rate of ventilation, the surface to volume ratio and the rate of particle placement of nested and unstick particles can also be affected. For example, a person's smoking causes a change in the concentration and the size of aerosol distribution and even the behavior of individuals is important. It is important that the smoker smokes a few cigarettes a day, and changes in the consumption patterns will lead to changes in parameters or changes in the household appliances, which can also be an important factor.

For this purpose, in this study, the concentration of particles of aerosols, the ventilation rate and the surface-to-volume ratio of the room as effective parameters were measured. These parameters were measured randomly in 13 houses and the results are shown in tab. 2.

In order to measure aerosol particle concentration particle counter machine of PC200-TROTEC model was used. This machine is used to control particulates in the air. It identifies aerosol particle concentration of 0.3, 0.5, 1.0, 2.5, 5, and 10 mm and displays them on 6.35 cm screen.

In this research, in order to determine the ventilation rate in a building, CO_2 was used as the indicator gas and TESTO535 machine was used to measure the CO_2 rate in the air. For this reason, before the injection of the indicator gas, the CO_2 concentration was measured. This machine measures the CO_2 rate with an accuracy of 1 ppm. By injecting the CO_2 into considered

Table 2. The average and the actual values of room parameters

Parameter	Min	Mean	Max
Aerosol particles connection [Z, cm^{-3}]	28000	64000	100000
Surface to volume room ratio [m^{-1}]	2	5	7
Air exchange rate [λ_V, h^{-1}]	0.1	0.5	2

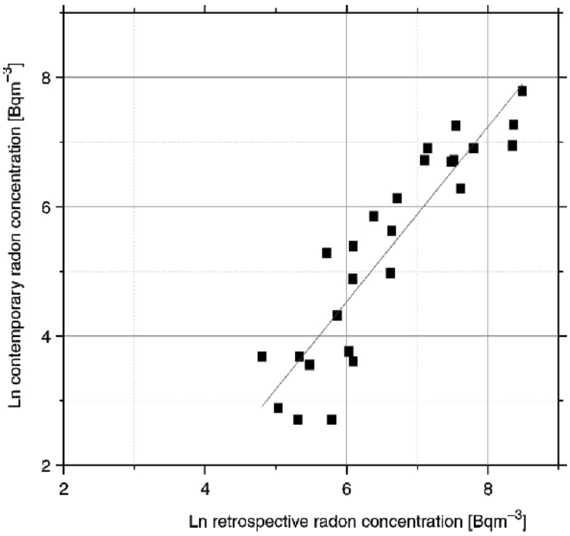

Figure 4. The comparison between the log of retrospective and the contemporary radon concentrations in Ramsar and Rasht residential homes

space and surveying the gas changes process, the gas changes rate per hour was calculated.

Also, to calculate the surface to volume ratio of the room, the area of each room and the volume of each room were obtained by taking into account the equipment in the room.

Based on the average global value provided by Bastrikov [30], after placing in eq. (2), the coefficient of 245 reached 212, which was not significantly altered. However, as noted, the change in room parameters over a period of 23 years can be one of the causes. The retrospective radon concentration was compared with the contemporary radon measurements as shown in fig. 4. The correlation coefficient between the retrospective and the contemporary radon gas concentrations is 0.80672.

The findings of this study are consistent with that of Zunic [33] which correlates well with the retrospective and the contemporary radon concentrations in 46 homes in Serbia. This correlation was also found by Bochicchio [6] in 21 objects in the homes of individuals that were non-smokers. However, in some studies, there was a weak correlation between the retrospective and the contemporary radon gas concentrations or no relationship was obtained between them [8, 34].

CONCLUSIONS

The present study provides a new approach for the use of ^{210}Po implanted surface activity to estimate the retrospective radon concentration. The glass surface activity was measured by Lexan PC as a solid state nuclear track detector to estimate the long-term average radon gas concentration for the first time. The surface trap technique, based on the Lexan PC detectors, was used in this study for the determination of ^{210}Po implanted in glass after the deposition of short-lived radon daughters. The surface activity measurements can provide reliable estimates of previous radon exposure. The activity of ^{210}Po implanted in glass surface was measured in different houses in Ramsar and Rasht cities of Iran. According to the obtained results, the relationship between the radon concentration based on ^{210}Po implanted in glass and the radon exposure by passive detector based on SSNTD are given by scatter plots. The results showed a moderate correlation between the contemporary and the retrospective radon concentrations. The average retrospective radon concentration was 2.4 times greater than that of the contemporary radon, and this could be due to the changes in the structure of the houses or living conditions of residents in these areas over a period of time. This assessment can be improved by measuring the parameters of the room, such as the air conditioning rate, the concentration of aerosols and the rate of stacking the particles stacked and nested more precisely. These results are useful in epidemiological studies which determine who may have been exposed to the variable radon concentration in their previous homes.

ACKNOWLEDGEMENT

This study was carried out within the frame work of a Ph. D thesis conducted at Department of Medical of Radiation Engineering, Science and Research Branch, Islamic Azad University, P. O. Box 14515-775, Tehran, Iran.

AUTHORS' CONTRIBUTIONS

The research idea was suggested by A. Azadbar and M. R. Kardan. The measurements were performed by A. Azadbar and S. Baradaran, and the analysis and the discussion were carried out by A. Azadbar, M. R. Kardan, D. Sardari, and S. Baradaran. The manuscript was prepared by A. Azadbar.

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Received on September 25, 2017
Accepted on April 16, 2018

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РЕТРОСПЕКТИВНО МЕРЕЊЕ РАДОНА НА ОСНОВУ ИМПЛАНТОВАНОГ ^{210}Po НА СТАКЛЕНИМ ОБЈЕКТИМА ПРИМЕНОМ ПОЛИКАРБОНАТНИХ ДЕТЕКТОРА

У циљу ретроспективног одређивања концентрације радона, извршена су мерења површински нанетог полонијума у 37 кућа у градовима Рашт и Рамсар у Ирану. CR-LR метода широко је примењивана за ове потребе, али је сада по први пут коришћен детектор од лексан поликарбоната за мерење активности ^{210}Po нанетог на стаклени објекат. Ови детектори су постављени на стакласту површину у периоду од 153 до 365 дана. Пасивна цилиндрична дифузиона комора је коришћена за мерења постојеће концентрације радона. Дифузиона комора се састоји од филмова лексан поликарбоната у улози чврстих нуклеарних траг детектора и филтера. Површинска концентрација ^{210}Po је измерена у опсегу 0.26 до 11.96 mBqcm^{-2} са средњом вредношћу од 2.62 mBqcm^{-2}. Осетљивост ^{210}Po у односу на поликарбонат износила је 0.06456 track cm^{-2} по mBqhcm^{-2}. На основу тога је одређена концентрација радона у опсегу од 122 до 4840 Bqm^{-3} са средњом вредношћу од 1243 Bqm^{-3}, а постојећа концентрација радона у околном простору износила је од 15 до 2420 Bqm^{-3}, са средњом вредношћу од 513 Bqm^{-3}. Резултати указују на постојање значајне корелације између ретроспективно одређене концентрације радона и егзистирајуће концентрације гаса радона у изабраним просторима са коефицијентом од 0.80672.

Кључне речи: стакласти објекат, ретроспективно мерење, ^{210}Po, поликарбонатни детектор, радон