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Detection of Radon Exhalation from Various Building Materials Using CR-39, RAD7, and Biosensors

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ABSTRACT

Study aimed to design and manufacture two biosensors, namely BIOS-I and BIOS-II, for ^{222}Rn and Pb+2 measurements in building materials and soil samples. For comparison, the conventional detectors of RAD7 and CR-39 were used. The biosensor material used was based on ssDNA rich guanine or primer. The two biosensors have a difference in the sequence of the nitrogenous bases. The measurement revealed that the average of ^{222}Rn exhalation by the BIOS-I was 373.30 Bqm-3, while the BIOS-II was 342.29 Bqm-3. The average ^{222}Rn exhalation measured by the CR-39 detector was 326.17 Bqm-3, whereas by the RAD7 detector it was 319.95 Bqm-3. This study found that ^{222}Rn exhalation in the Indian and Chinese granites, soil, and Iraqi mosaic samples was higher than the limits recommended by WHO, while the rest of the samples were within the permissible limits. It is also known that there is a very weak positive correlation between BIOS-I or BIOS-II and humidity, while a very weak negative correlation was found between them and temperature. There is a very strong positive correlation between radon exhalation recorded by RAD7 and humidity. On the other hand, there are no statistically significant differences between BIOS-I and BIOS-II at (level 0.01), while there are statistically significant differences between BIOS-I and CR-39 or RAD7 at level 0.01. It was concluded that the manufactured biosensors have better detection for radon than RAD 7 and CR-39 detectors.

INTRODUCTION

Many toxic compounds such as organic pesticides, heavy metals, and other pollutants can be found in various environments, and cause human health problems such as cancer, endocrine-disrupting activities, acute congestion, and degenerative changes in the nervous systems, as well as environmental problems [1]. Therefore, it is necessary to develop biosensor technologies for continuous monitoring in environmental and healthcare applications that can provide on-site, real-time detection and quantification of beneficial and toxic compounds, such as at the laboratory bench, in the field, or in storage room facilities [2]. A biosensor is a device that uses specific biochemical reactions mediated by isolated enzymes, immune systems, tissues, organelles or whole cells to detect chemical compounds usually by electrical, thermal or optical signals. All biosensors are based on a two-component system: (a) biological identifier elements (ligand) that facilitate specific binding or biochemical reaction with the target analytes, and (b) Signal conversion units (transducers) [3].

In general, the biosensors can be categorized into two main groups: (1) sensors that can directly recognize a target substance (i.e., directly measure the biological interaction), and (2) sensors that can indirectly recognize a target substance. This type relies on secondary elements to detect a substance (usually using catalyst such as fluorescent tags or enzymes).

Radon is a radioactive noble gas commonly found in soil, water, and materials used in construction and interior decorating [4,5]. Radon enters the human body through inhalation, and it

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causes internal injuries by way of radiation. The World Health Organization (WHO) classifies radon as one of 19 environmental carcinogens [6]. The International Agency for Research on Cancer (IARC) confirmed radon and its decay products as carcinogens representing the second highest risk factor for lung cancer [7]. These warning reflects the widespread concern about the threat radon poses to human health [8]. Therefore, the detection of radon is a significant task.

For many years, instruments for the measurements of radon and its decay products in research were based mostly on the detection of alpha particles. The radon concentration measurement based on sampling methods can be divided into three categories viz. instantaneous sampling, continuous sampling and cumulative sampling [9]. However, the conventional method of measuring accumulated radon radiation is greatly affected by climatic factors like temperature and humidity, and the measurement procedures are complicated and time consuming. Furthermore, the on-site sampling and measurement process exposes operational personnel to radiation. Even in a laboratory-environment, it is highly hazardous to perform measurements involving radioactive materials. Over the years, scientists have developed many new methods of detecting radon [10-13].

The aim of this study was to design and manufacture two biosensors for ^{222}Rn and Pb^{+2} exhalation measurements in some building materials and soil samples. This is new method for detecting ^{222}Rn and Pb^{+2} in some materials in Iraq.

METHODOLOGY

The samples consist of building materials and soil. The building materials were taken from the local markets in Basra City while the soil samples were taken from three different sites in the Al-Midaina district using GPS, as shown in Tables 1 and 2.

The collected 18 samples were crushed into small pieces, then converted to a fine powder using a manual grinding and an electrical mixer (Sinico, Germany), as shown in Fig. 1. The fine powder was sieved to obtain grain size of 300 μm for about 500 g in weight using special sieves mesh. Then, the samples were dried at 100°C for 2 hours using an oven (Model Memmert GmbH+ Co. KG, Germany). The prepared powders were then packaged in a moisture-proof food bags and stored for about one week prior to measurements to achieve the secular equilibrium for ^{222}Ra and their progenies [14]. The two manufactured (Biosensor-I and biosensor-II) have a sequence as follow: 5′-AGGGTTAGGGTTAGGGTTAGGG-′3) and 5′-GGTTGGTGTGGTTGG-′3, respectively.

Table 1. Code, type and country origin of samples detected.

Sample No.	Sample code	Type	Country of Origin
1.	G1	Granite	India
2.	G2	Granite	China
3.	C1	Ceramic	India
4.	C2	Ceramic	Iran
5.	Mo1	Mosaic	Iraq
6.	Mo2	Mosaic	Iran
7.	M1	Marble	Iran
8.	M2	Marble	Turkey
9.	B1	Brick	Iran
10.	B2	Brick	Iraq
11.	T1	Thermostone	Iran
12.	T2	Thermostone	Iraq
13.	Ce1	Cement	Iraq
14.	Ce3	Cement	Iraq
15.	Loc1	Soil	Iraq/Basrah
16.	Loc2	Soil	Iraq/Basrah
17.	Loc3	Soil	Iraq/Basrah

Table 2. Coordinates of sampling sites.

Code	Longitude	Latitude
Loc1	47º 15′ 38″	31º 00′ 12″
Loc2	47º 15′ 08″	30º 58′ 27″
Loc3	47º 18′ 14″	30º 58′ 11″

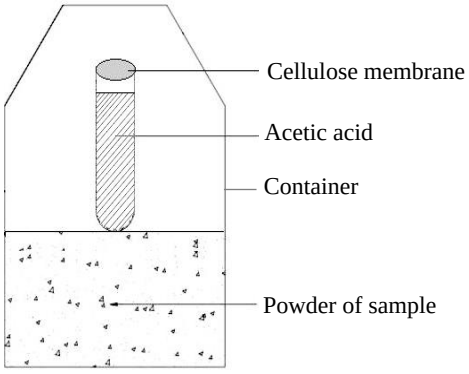


Fig. 1. Illustration of the biosensor cell that consists of cellulose acid in a test tube and sample powder in a bottle.

To make a biosensor cell, the sample was placed in a 100 g plastic container and a tube filled with 10 ml of 0.2 % acetic acid was partially placed in the same container. The tube was covered with a cellulose acetate membrane (Sartorius Stedim Biotech, Germany) to prevent other contaminants from entering the acid. The pore size of the

cellulose acetate membrane is about 45 µm, as shown in Fig. 2.

The cell was placed in a vacuum chamber where it was connected to a vacuum pump (JB Industries, USA) equipped with a pressure gauge, as shown in Fig. 2. The cell (acid and sample) remained under a pressure of -50 bar for 8 days inside the vacuum in order to obtain a large amount of radon gas from the sample. After the end of 8 days' exposure time, two milliliters of acid were added and transferred to two containers, then 20 µl of aptamer was added in each container at a concentration of 0.6µM and then the containers were placed in an incubator at 37^{o}C for 90 minutes. Then, 22 µl of malachite green dye (AVONCHEM, UK) was added and put back into the incubator at the same temperature for 15 minutes. Mixture of acid and the primer stimulated a conformational change of aptamer resulting in G-quadruplex, which is a characteristic of guanine-rich primers. Malachite green is a crystalline green dye with a metallic sheen having three methyl groups [15]. For strong fluorescence, there must be an interaction between green malachite and G-quadruplex [2]. Uda et al. (2017) reported that the fluorescence spectroscopy for energy transfer from green malachite can identify single and double-stranded DNA and G-quadruplex in molecules [16]. The presence of green malachite and G-quadruplex together in the same group gives a strong fluorescence. The test under optimal condition showed the best dye volume at 22 µL. This technique is characterized by its ability to provide accurate detection of radon gas and lead ion by providing very high fluorescence as can be seen in Fig. 2 [17].

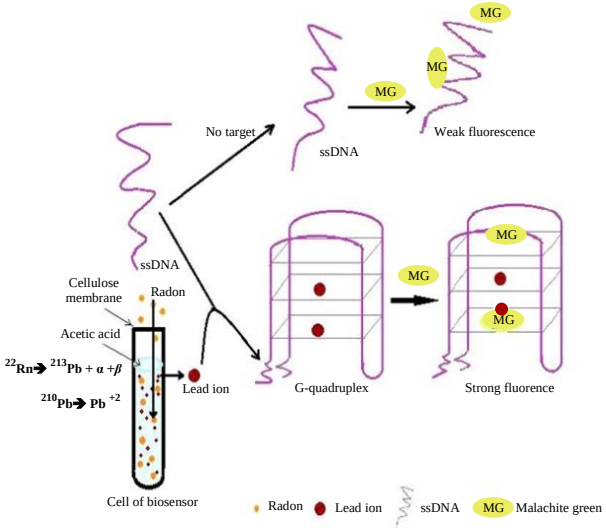


Fig. 2. Illustration of the G-quadruplex.

The building material samples were collected from different area in Iran, Iraq, and China as listed in Table 1. These samples were grounded, dried and screened using a special sieve with grain size of 300 µm. Each sample was weighed 100g and placed in a plastic cup. The CR-39 track detector pieces was mounted on the top of the plastic cup, where the sample was placed in the plastic cup, and closed with a stopper.

The chemical solution has the ability to affect the damaged areas by alpha particles [18]. The chemical solution was prepared by dissolving 50 g of NaOH (6.25 N) in 200 ml of deionized water. The weight of the solution can be determined using the Eq. (1).

$$W = Weq \times N \times V \quad (1)$$

where:

W = NaOH weight (50 g)
Weq = Equivalent mass of NaOH (40 g)
N = Normality (6.25 N).
V = Volume of distilled water (200 ml).

After 72 days of exposure, a chemical etching process was carried out by immersing the CR-39 detector in a beaker containing the chemical solution. Then, the solution with the detector CR-39 was heated up for 6 hours at 75 °C using a water bath (GFL, 1083, Germany). In this step, the beaker was sealed tightly to prevent the evaporation of solution and any changes in NaOH concentrations during the heating process. After chemical etching process, the detectors were washed with a tap and deionized water, then dried with a filter paper for 15 minutes.

The tracks of alpha particles were counted in CR-39 detectors using an optical microscope (KRUSS, OPTRONIG, Germany) at 10× amplification. The microscope was equipped with a camera (MDCE-5C) for displaying the tracks on a computer screen.

To obtain the calibration factor (1/s), three dosimeters had been prepared in a constant volume of plastic containers. First detector was exposed by ^{226}Ra for 3.83 days, while second and third detectors were exposed, respectively for 5 days and 6 days. The radon exhalation was measured by RAD7 as shown in Fig. 3.

Fig. 3. RAD7 system for measuring radon concentration.

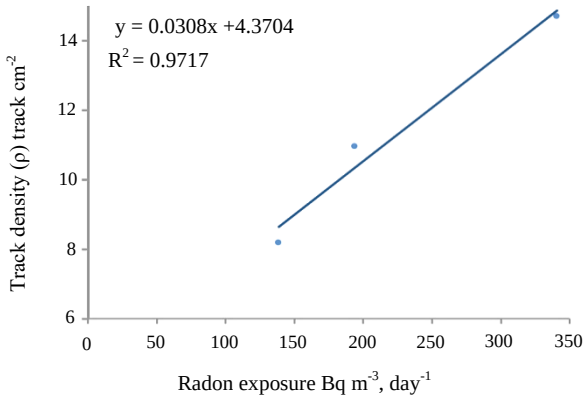


Fig. 4. A typical calibration curves for radon exposure for the CR-39 detector.

Radon gas concentration was measured for 17 different samples of building materials. Each sample was weighed of 100 g and placed inside the vacuum system. The samples were vacuumed to a pressure of about -50 bar, then placed for 8 days and examined by RAD7 device (Durridge company, USA). The settings of the device were in the sniff mode with eight cycles for 10 minutes each.

It is important to note that the etching procedures are the same as above. The measured calibration factor (1/s) was 0.0308 track.cm^{-2}/Bq.m^{3}. d, with a standard deviation of ±1.99. A typical calibration curve for CR-39 detector is shown in Fig. 4.

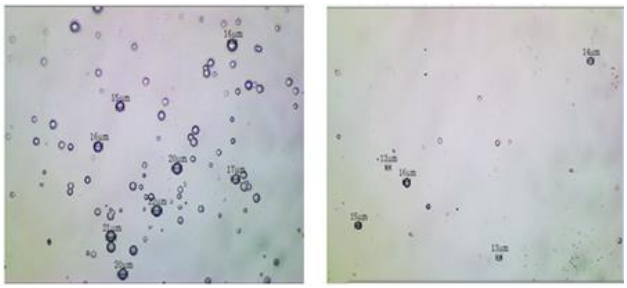
RESULTS AND DISCUSSION

Figure 6 shows the results of measurements of radon exhalation (^{222}Rn) using BIOS-I. The sensor consists of a primer that contains 12 nitrogenous bases of guanine type. The radon exhalation using BIOS-I was found to be in the range between 138.55 Bqm^{-3} and 735.40 Bqm^{-3}. The lowest radon gas exhalation was 138.55 Bqm^{-3} in the Iranian marble, whereas the highest radon exhalation was 735.40 Bqm^{-3} in the Indian granite.

Figure 6 shows the radon gas exhalation measured by BIOS-II. It consists of a primer that contains 9 nitrogenous bases of guanine type. The ^{222}Rn exhalation measured by BIOS-II found to be in the range from 116.12 Bqm^{-3} to 705.64 Bqm^{-3}. The lowest radon exhalation was 116.12 Bqm^{-3} in the Iranian marble, whereas the highest radon exhalation was 705.64 Bqm^{-3} in Soil Loc2. The highest ^{222}Rn exhalation in Chinese granite, soil (Loc2), and Indian granite may be due to the nature of the components involved in the formation of granite that form underground igneous rocks and under high temperatures. It contains mainly feldspar, quartz, mica and amphibole minerals in its components. It is noted that the granite samples have a high level of the radon gas because these elements melt with lava and are re-formed. The availability of radioactive materials other than Radon is ^{137}Cs that is found in agricultural and residential. The lowest radon exhalation from the marble may be due to the geological nature of the mountains where marble is taken from metamorphic rocks comprising a crystalline aggregate of calcite and/or dolomite in its basic components. This material is characterized by its low radioactivity while ceramics have low releasing of 222Rn because the raw materials for ceramics belong to the earth's crust, which contains naturally occurring radioactive materials in trace amounts. The cement could be a fine powder of calcium alum inosilicate which is made by pulverizing a blend of limestone and clay and heated up to almost 815 ºC. The average measurement results by using BIOS-I was 373.30±18.66 Bqm^{-3} while by using the BIOS-II was 342.29±17.11 Bqm^{-3}. The values recorded by the BIOS-I were slightly higher than the values recorded by the BIOS-II because of the primer used to manufacture the BIOS-I contains guanine nitrogenous base by 12 more bases than the primer used in BIOS-II. It contains 9 nitrogenous bases of guanine type. This difference in nitrogenous bases gave BIOS-I high accuracy because it increased the ability of Biosensor1 to stimulate conformational changes in order to create the selective advantage of lead and radon (G-quadruplex). Table 3 shows the Pearson correlation between ^{222}Rn and humidity and temperature with a weak correlation. There is a good correlation found between radon BIOS-I and BIOS-II while there is a weak correlation found between all types of biosensor with temperature. There is a very weak positive correlation found between all types of biosensor with humidity. The correlation shows one of the most important and strongest properties of biosensor [19,20]. It is not affected by humidity and temperature compared to the rest of the sensitive to humidity, temperature, light, and others. This feature can be used to obtain results with high accuracy compared to the conventional methods.

Table 3. Pearson correlation between biosensor, humidity and temperature.

Pearson Correlation N=17	^{222}Rn BIOS-I	^{222}Rn BIOS-II	RH	T
^{222}Rn BIOS-I	1	0.999**	0.025	-
^{222}Rn BIOS-II	0.999**	1	0.041	-
RH	0.025	0.041	1	0.6
T	-0.122	-0.104	0.630**	1



(a) (b)

Fig. 5. Tracks diameter of high (a) and low (b) of alpha tracks of Indian granite.

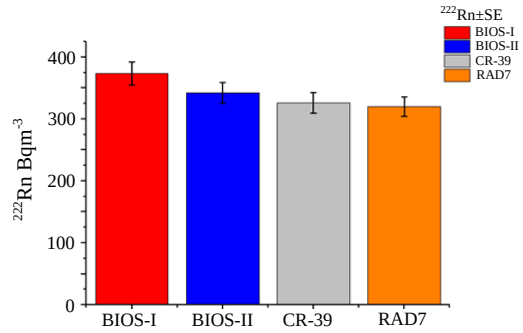


Fig. 6. Comparison of measurement results of average ^{222}Rn by four different detectors.

Figure 5 shows the tracks diameter distribution for Indian granite with an etching time of 6 h. The average diameter value of tracks for Indian granite was 16.69 ± 0.87 μm.

The value of radon exhalation measured by CR-39 was found to be ranging from 88.61 Bqm-3 to 702.57 Bqm^{-3}. The lowest radon gas exhalation was 88.61 Bqm-3 from the Iranian.

Perhaps it was due to the humidity or the influence of temperature. The average of radon gas exhalation was 326.17±17.05 Bqm^{-3}. The value of radon by RAD7 was found to be ranging from 82.47 Bqm^{-3} to 683.93 Bqm^{-3} with an average of about 319.95±15.99 Bqm^{-3}. The lowest radon gas was 82.47 Bqm^{-3} from the Iranian marble, whereas the highest radon exhalation was 683.93 Bqm^{-3} from the Indian granite.

Figure 6 shows a comparison of average ^{222}Rn of four different detectors. The mean radon recorded by BIOS-I was 373.30 ± 18.66, while the mean radon recorded by using BIOS-II was 342.29 ± 17.11 Bqm^{-3}. It appears that the values of radon gas recorded by CR-39 and RAD7 were lower than those recorded by BIOS-I and BIOS-II. They were at an average of about 326.17±17.05 and 319.95±15.99 Bqm^{-3}, respectively. Perhaps, this variation in the results is due to the high values recorded by the biosensor because it is a type of detector that is not affected by heat, humidity, light, etc., as it gives constant and accurate values. The radon gas exhalation recorded by CR-39 and RAD7 are much lower because these types of detectors are affected directly by humidity and less by heat, or in case of CR-39, it depends on the chemical etching process.

There is a good positive correlation found between radon (RAD7) and humidity, while there is a weak negative correlation found between radon (RAD7) and temperature. Perhaps the strong association between radon (RAD7) and humidity can be explained because the efficiency of the RAD-7 is dependent on the humidity of the chamber. A high humidity causes a smaller counting rate.

Table 4. Statistical significant differences among BIOS-I, BIOS-II, CR-39, and RAD7.

Sig. (2-tailed) N=17	^{222}Rn BIOS-I	^{222}Rn BIOS-II	CR-39	RAD7
^{222}Rn BIOS-I	….	0.013	0.554	.757
^{222}Rn BIOS-II	.013	….	0.393	.559
CR-39	.544	.393	….	.255
RAD	.122	.559	.255	….

Table 4 shows statistical significant differences among biosensors, CR-39, and RAD7 at level 0.01. There are no statistical significant differences found between BIOS-I and BIOS-II at level 0.01 while there are statistical significant differences found among BIOS-I, CR-39, and RAD7 at level 0.01. It was weak negative correlation between all types of biosensor with temperature. And it was very weak positive correlation between all types of biosensor with humidity. There was a very strong positive correlation between RAD7 detector and humidity. There are no statistical significant differences found among CR-39 with BIOS-I and RAD7 at level 0.01.

CONCLUSION

The manufactured biosensors can be used to detect radon gas exhalation and depend on the aptamer that is rich in guanine bases. The sensitivity of the biosensor in the first primer is higher than the second primer because the first primer is rich in base guanine. The G-quadruplex property is highly sensitive to radon gas. The BIOS-I is more sensitive than BIOS-II for radon exhalation measurement. It was found that ^{222}Rn concentration in Indian and Chinese granites, soil, and Iraqi mosaic samples were higher than the limits recommended by WHO (100 Bq m-3). The rest of the samples were within the permissible limits by WHO. Granite and soil are significant sources of radon gas. It is concluded that the biosensors have better radon detection than the RAD 7 and CR-39 detectors.

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AUTHOR CONTRIBUTION

Basim Almayahi: Conceived and designed the experiments; Performed the experiments; Analyzed and interpreted the data. H. J. Albazoni: Contributed reagents, materials, analysis tools or data; Wrote the paper.

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