

Research Article

Application of the Electret in Alpha Radiation Sensor to Measure the Concentration of Radon in Selected Ambient Conditions

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A novel electret sensor, made from a polytetrafluoroethylene (PTFE) foil, is proposed for monitoring of radon concentration in closed spaces. A triode air system was implemented to form the electret foil. In order to calibrate the electret sensor, the ionization chamber with the electret embedded at its bottom, was placed in an emanation chamber containing radon of known concentration. The change in the effective charge density of the electret resulting from the interaction of the electret sensor with ionizing radiation, make it possible to determine the degree of its response to the radon concentration in the examined environment. The electric charge of the electret sensor was evaluated at the beginning and at the end of the concentration measurement using a field chopper. The measurement of the value of the electric field between the electret and the reference electrode directly corresponds to the effective surface charge density of the electret. The measuring system was calibrated in values of the electret equivalent voltage of U_z . In order to assess the electret properties, both the thermostimulated charge relaxation (TSQ) method and the thermostimulated discharge current (TSDC) method were used to evaluate corresponding parameters. These investigations led to the optimization of the electret formation process for the maximum sensitivity to the effects of alpha particles resulting from radon radioactive decay.

1. Introduction

Radon, a naturally occurring noble gas, is the main source of ionizing radiation in the human environment. On average, the concentration of radon in enclosed spaces utilized by people is several times greater than that in the outside air. In order to prevent health risks associated with exposure to this radioactive element, it is necessary to monitor its concentration in enclosed spaces. Monitoring radon concentration using a passive dosimeter, made from a polytetrafluoroethylene (PTFE) foil, as well as the assessment of the effect of air humidity on the obtained results of concentration measurements in the investigated space have been carried out earlier [1]. A warning system, designed to signal when the amount of ionizing radiation exceeds the permissible concentration limit, ensures human safety in such spaces [2–5].

An accurate determination of the annual average concentration of radon requires that it be measured for a period of at least two months. In order to measure the instantaneous concentration of this radioactive gas, the minimum measurement time should be at least 48 hours. In general, the longer the measurement time the more accurate and reliable are the results obtained [3–7].

1.1. Radiation Hazard. The following aspects are the reasons for the hazardous impact of radon Rn222 on human beings.

- (i) Being the most stable, statistically radon is the main source of human exposure to radiation
- (ii) Radon is the only gaseous radioactive element and can migrate from the rocks and soil to the earth's surface

- (iii) Radon decay products (polonium, bismuth, lead) are not chemically inert and can be embedded in lung tissue where they will stay for an extended period of time eventually destroying the tissue. For example, if the decay of Rn222 occurs in the airways, the solid Po218 produced can become embedded in the bronchial or alveoli walls. The exposure of the respiratory system to radiation associated with its decay chain can cause lung cancer. If the decay of Rn222 happens outside the body, the decay products may attach themselves to the solid particles present in air forming so called radioactive aerosol, which can enter the airways with inhaled air and become partially deposited in the airways
- (iv) Statistically, radon is more dangerous than other chemicals, as it causes much higher mortality than other carcinogenic substances. Compared with asbestos, known as a potent carcinogen, radon causes 10 times more deaths
- (v) The concentration of radon indoors is on average several times higher than in the open air, which is especially dangerous for modern societies in which people spend a larger portion of their lives inside buildings [1–5].

1.2. The Migration of Radon into Confined Spaces. As the only gaseous radioactive element, radon migrates from rocks and soil into buildings through all possible leaks in the buildings' foundations. It can get into occupied spaces through fissures in both incorrectly assembled ventilation ducts and utility installation (such as water and drain plumbing) conduits. In most countries, there are norms that specify the maximum average concentration of radon inside residential and office buildings. For example, radioactivity limit is 800 Bq/m³ in Scandinavian countries, 250 Bq/m³ in Germany, and 150 Bq/m³ in the USA. In Poland, consistent with the recommendations of the National Atomic Energy Agency, the acceptable average concentration of radon in new buildings (completed after 01.01.1988), should not result in radioactivity exceeding 200 Bq/m³ [1]. In older buildings, the average radioactivity should not exceed 400 Bq/m³. The World Health Organization recommends that the average annual radon activity in human environment should not exceed 100 Bq/m³. According to the EU directive, the radioactivity limit is set at 300 Bq/m³ [7–11].

1.3. Methods of Measuring the Concentration of Radon and the Products of Its Decay. The commonly used methods to determine radon concentration in air confined in closed spaces can be classified as direct or indirect. The direct methods are characterized by the fact that the concentration measurement takes place on site using specialized dosimeters. Indirect methods require two steps. In the first step, a detector containing either active carbon adsorbing radon or a special foil sensitive to alpha radiation is placed in the investigated space. The second stage involves the analysis of the detector state in a specialized laboratory.

Considering the measurement time, the methods can also be classified as short term, long term or continuous. In short term methods, the detector is exposed to the radioactive factor for a period from 2 to 7 days. In long-term methods, the detector exposure period varies from 1 to 12 months. In continuous methods, the measuring instrument always updates the average value of the radon concentration over a predetermined time period (for example 10 min, 30 min, etc.) [8].

1.4. Techniques for Measuring the Concentration of Radon. Listed below are the most common methods of measuring radon concentration:

- (i) Observation of alpha particle traces in a chamber with nuclear emulsion
- (ii) Scintillation measurements using a Lucas cell
- (iii) Measurements using activated charcoal and gamma spectroscopy

Currently, there is a growing interest in miniaturized, portable systems with the option of real time data collection. These systems can often be controlled through the Internet making direct and continuous measurements possible. The most common solutions are listed below.

- (iv) Continuous recording on a filter [9] - These measurements consist of analysis of radon-containing air by passing it through a filter impervious to radon decay products. The measurement of the activity of decomposition products embedded during the test on the filter make it possible to assess the concentration of radon in the studied air
- (v) Measurements using an integrated electret radon meter [2–7] - The change in the charge or the corresponding equivalent voltage of the electret constituting the radon sensor is assessed during the measurements. Depending on the concentration of radon in the studied environment, the sensor responds with a greater or smaller drop in the equivalent voltage. The values of the equivalent electret voltage at the beginning and the end of the exposure provide the concentration of radon

1.5. Electret Sensors for Measuring Radon Concentration. As with all dielectrics with an externally created metastable electric polarization, electrets can be used not only to measure the concentration of radon but also to detect beta and gamma radiation in confined spaces. When the detector is subjected to radiation, the ionization of the air in the chamber causes compensation of the electret's polarization charge. This property makes the electrets feasible for alpha, beta, and gamma dosimetry. Studies have shown that the difference in uncompensated electric charge, measured before and after irradiation, is a linear function of the radiation dose. The slope of the line, representing the change in the uncompensated charge as a function of the electret exposure time to the radiation, defines the sensitivity of the dosimeter.

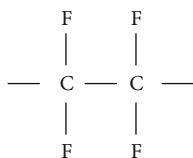


FIGURE 1: The structural formula of polytetrafluoroethylene.

A passive method of measuring radon concentration indoors and outdoors is described in [6]. Ionization chambers containing an electret, whose electric charge is insignificantly dependent on external factors, such as temperature and humidity, are also suitable for outdoor conditions. These devices, often referred as Radon Monitors, have an electret ionization chamber (EIC) design and are known as electret-passive environmental monitors (E-PERM). Measurements of radon concentration in the environment using a passive measurement method are also described in [7–11].

2. Experimental Details

2.1. Forming of an Electret from a Polymer Film. A polytetrafluoroethylene (PTFE) foil has been selected for preparation of electrets intended for use in the designed radon dosimeter. The polymer structural formula is shown in Figure 1.

A dosimeter, in which an electret is used for radon detection, makes it possible to assess its concentration in enclosed spaces by performing investigations lasting from a few days to two weeks. The principle of dosimeter operation is based on the property that the radiation resulting from radon decay compensates the electrical charge introduced earlier in the electret volume. Alpha particles, formed in the ionization chamber during the radioactive decay of radon, are indirectly responsible for the compensation of the electret charge. Immediately after decay, emitted alpha particles with an energy of about 6.5 MeV, ionize the air contained in the electret's vicinity. The negative ions, formed as a result of this process, compensate the positive charge of the electret. Comparison of the electret charge, or the corresponding equivalent voltage of the electret, measured just before and immediately after the exposure to radiation makes it possible to evaluate the concentration of the radiation source in the investigated environment [2–5].

The use of dosimeters for short-term and long-term investigations requires electrets with various sensitivities. This can be achieved by the application of different materials as well as different thicknesses of the foils used for the electret inserts in the dosimeters. In the prototype dosimeter, the electret inserts were made from a PTFE foil in the form of flat, circular wafers 35 mm in diameter and with a thickness of 0.5 mm. Properly selected inserts were placed at the bottom of a specially constructed ionization chamber. The air containing radon diffused into the ionization chamber through a filter which eliminated the products of an earlier radon decay. For the correct evaluation of radon concentration, air containing only radon enters the ionization chamber [6, 7].

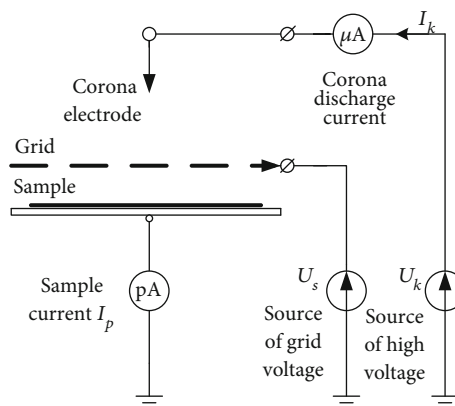


FIGURE 2: The corona discharge system for the fabrication of electrets [12].

A corona discharge system configured in a triode air arrangement was utilized for forming electrets from the PTFE foil [10, 12]. This solution made it possible to obtain electrets with a consistently reproducible value of the introduced charge (equivalent initial voltage of the electret). In the process, a high-density charge is implanted into the polymer film structure. The applied method guarantees that the electrets thus obtained will be long lived and have a stable value of the effective charge density accumulated in the electret volume (the equivalent voltage) [12]. The setup for fabrication of the electrets at room or elevated temperatures is shown in Figure 2.

The system contained a discharge electrode placed above the electret sample, a metallic grid supported on a ceramic ring holding the sample, and a conductive electrode constituting the base for the processed foil sample. The conductive electrode was rigidly fixed in the system while separated galvanically from other conductive elements. The corona electrode was made as a 10 mm long needle with a tip radius of approx. 50 μm . The corona electrode was mounted at a distance of 45 mm from the foil sample used for fabrication of an electret. The third electrode of the system, the grid, was fixed permanently at a distance of about 9 mm above the sample. The metallized on one-sided foil sample, used for electret formation, was placed under the grid on the lower electrode grounded through a picoammeter. The sample was pressed against the electrode with a ceramic ring. The following forming conditions were adopted: corona voltage $U_k = +12$ kV, corona current $I_k = 5$ μA , the grid voltage was determined $U_s = +1400$ V.

Electret formation began when simultaneously both the grid supply voltage and the corona discharge voltage were turned on. From the moment the voltage was switched on the current flowing through the sample I_p was measured [12]. The corona discharge system was placed in a chamber with controlled temperature. The power of the chamber heater was 470 W and a PT 100 resistance temperature detector was used to control the temperature inside the chamber. The detector was placed in a hole drilled in the lower electrode of the system directly under the sample. The applied solution makes it possible to continuously

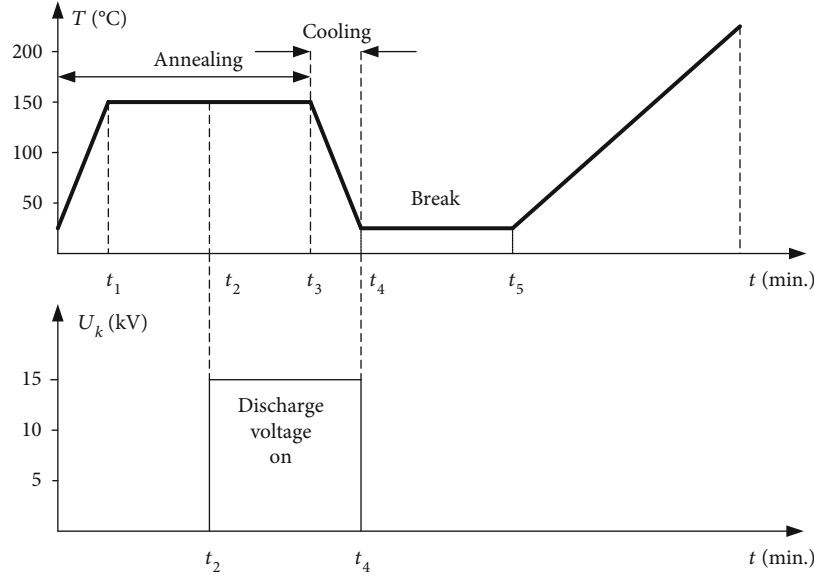


FIGURE 3: Changes of processing temperature and corona voltage during the formation (from t_2 until t_4) and investigation of the electret (after t_5).

control the temperature of the formed electret. The programmed changes in temperature inside the chamber, necessary in the electret fabrication process, were carried out using a programmable temperature controller constructed for this purpose. This solution made it possible to form foil electrets at ambient temperature as well as at temperatures of 100° C and 150° C.

Time dependency of temperature during the formation process and the electret investigations together with the time of turning on and off the high voltage supply is shown in Figure 3.

After formation and a 24 hour break, the electret lifetime was determined from the analysis of the curves obtained during the process of thermo stimulated relaxation of the electret by monitoring either its charge (TSQ) [13] or its discharge current (TSDC) [13–15].

2.2. Determination of the Electret Parameters Using the TSQ Method. The time during which the electric charge is retained, often referred to as the electret lifetime, in a formed foil electret determines its suitability for practical applications. In this experiment, the electret lifetime was determined from the analysis of the curves obtained in the process of thermo stimulated discharge [13]. The lifetime was found from the TSQ spectrum by analyzing the slope of the secant obtained from the points, representing the values of the equivalent voltage U_z for selected temperatures T_n and T_e (its slope at the inflection point). The equivalent voltage of the sample was monitored using the voltage compensation method [10, 16]. The measurements were conducted in conditions under which the temperature of the electret samples was linearly raised. The obtained spectrum, such as the one shown in Figure 4, led to the determination of the activation energy in the relaxation process of the studied electret.

In the single relaxation process, the change in in the electret charge is described by the exponential function [13]:

$$q_s(T) \approx q_{s0} \exp \left[-\frac{kT^2}{bW\tau_0} \exp \left(-\frac{W}{kT} \right) \right] \quad (1)$$

where q_s and q_{s0} represent the effective surface charge density at temperature T and its initial value after implantation, b is the sample heating rate during the experiment, W stands for the activation energy in the relaxation process, k is the Boltzmann constant, and τ_0 a time constant.

The activation energy was evaluated from Equation (2).

$$W = -\frac{kT_n^2}{U_z(T_n)} \text{tg}\alpha \quad (2)$$

in which W represents the activation energy of the relaxation process, T_n corresponds to the temperature at the inflection point of the TSQ spectrum, $U_z(T_n)$ denotes the value of the equivalent voltage at the inflection point (corresponding to the effective surface charge density of the electret at temperature T_n), $\text{tg}\alpha(T_n)$ stands for the slope of the curve at the inflection point, and k is the Boltzmann constant.

Finally, the electret lifetime was found from Equation (3) [13].

$$\tau(T) \approx \frac{kT_e^2}{bW} \exp \left[\frac{W}{k} \left(\frac{1}{T} - \frac{1}{T_e} \right) \right] \quad (3)$$

where $\tau(T)$ is the electret lifetime at temperature T , T_e stands for the temperature at which the equivalent voltage has dropped by a factor e from its maximum value ($U_{z \max}/e$), W represents the activation energy of the relaxation process, b is the sample heating rate.

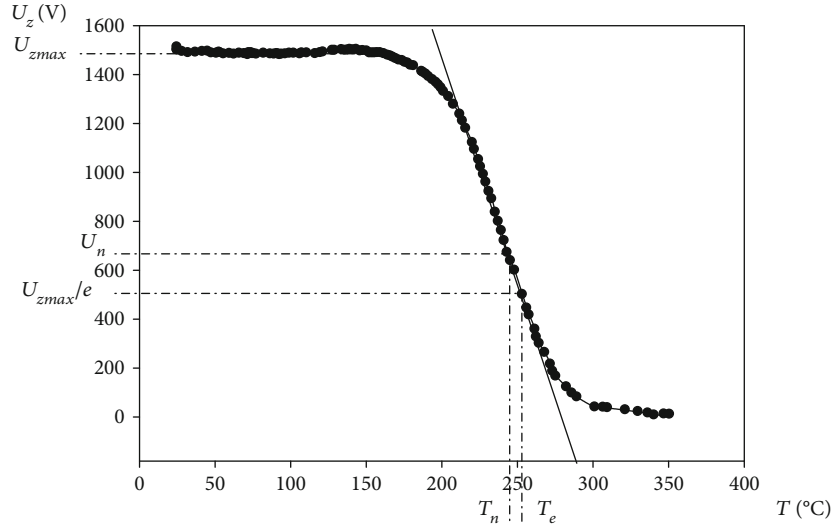


FIGURE 4: The TSQ spectrum for a sample prepared at room temperature.

TABLE 1: The spectrum parameters, activation energy and the lifetime obtained in the TSQ investigations of an electret sample shown in Figure 4.

B [K/s]	U_{zmax} [V]	U_{zmax}/e [V]	T_e [K]	U_n [V]	T_n [K]	$Tg\alpha_{(T=T_n)}$	W [eV]	$\tau(300\text{ K})$ [s]
0.033	1500	553.5	526	700	521	18.91	0.624	3.6×10^7

The characteristic parameters of the TSQ spectrum, shown in Figure 4, as well as the calculated activation energy and lifetime, are given in Table 1.

2.3. Determination of the Electret Parameters Using the TSDC Method. In order to verify the TSQ results described earlier, the lifetime of the electret charge was also determined by an alternative, thermally stimulated depolarization method. In the TSDC method, an electrometer is used to measure the depolarization current in a closed circuit, containing the sample with deposited metallic electrodes. The study is also carried out at a linearly increasing temperature.

As a result of the current measurement, we obtain a characteristic relation referred to as the TSDC spectrum. The spectrum consists of isolated or overlapping current peaks. Analysis of such a spectrum is complicated due to the mutual overlap of relaxation processes, and the separation of individual peaks is difficult and usually encumbered with a large error. In order to estimate the relaxation parameters of individual processes, genetic algorithms were implemented [13, 14]. The TSDC spectrum of a studied electret film, which results from a superposition of two relaxation processes α and β , is shown in Figure 5.

Using the characteristic points of the high temperature peak β in the TSDC spectrum, the first order estimate of the activation energy of the relaxation mechanisms were determined from the following relation:

$$W = \frac{0.976kT_m^2}{T_2 - T_m} \quad (4)$$

in which W is the activation energy corresponding to a particular relaxation mechanism, k – the Boltzmann constant, T_2 – the temperature, where the spectrum reaches half of the maximum current on the higher temperature side, T_m – the temperature at the corresponding maximum is the spectrum [10].

The parameters of the relaxation processes thus obtained, associated with the overlapping current peaks, were used as the input values for the analysis of TSDC spectra using a genetic algorithm. The calculations implemented the Matlab Global Optimization Toolbox library [15, 17]. The library contains specialized genetic functions for both binary and floating point representations. Based on the library resources, an application for analysis of TSDC spectra using the genetic method was built. The application requires defining an objective function, determining the search ranges of individual parameters, a criterion for completing the calculations, determining the size of the population, choosing the type of selection, mutation and crossover as well as their parameters. This method leads to the separation of individual relaxation peaks even from a complex TSDC spectrum [15, 17]. In Figure 6, the plots corresponding to individual relaxation mechanisms are identified, as well as the result of their superposition, which is compared with the collected data.

The optimal parameter values for the activation energies W, effective surface charge densities P_0 , initial time constants τ_0 , and the electret lifetimes τ , obtained in the genetic algorithm, for the isolated relaxation processes α and β , identified in the electret foil, are listed in Table 2. From the activation energy, found by using the genetic algorithm, the values of the charge lifetime have been determined for both observed relaxation mechanisms as $\tau_1 = 7.71 \cdot 10^5$ s and $\tau_2 = 7.9 \cdot 10^5$ s.

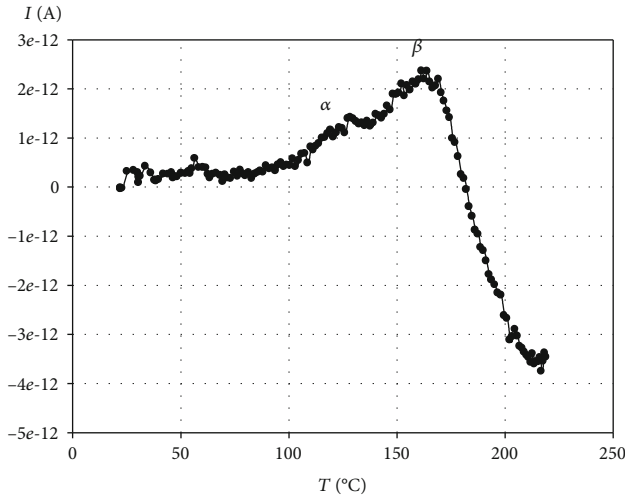


FIGURE 5: The TSDC spectrum in an electret sample.

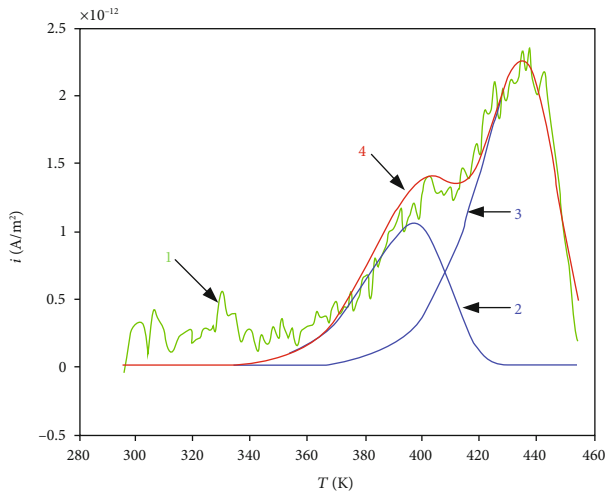


FIGURE 6: The analysis of the positive part of the TSDC spectrum (curve 1) based on the genetic algorithm for two isolated relaxation processes (curve 2 and 3) and their superposition (curve 4) as the fit to the experimental curve.

TABLE 2: Relaxation parameters obtained from the genetic algorithm.

	Peak 1	Peak 2
W [eV]	0.936	1.157
P_o [C/cm ²]	$8.68 \cdot 10^{-10}$	$1.82 \cdot 10^{-9}$
τ_o [s]	$4.35 \cdot 10^{-10}$	$1.24 \cdot 10^{-11}$
τ [s]	$7.61 \cdot 10^5$	$7.9 \cdot 10^7$

3. Application of Electrets for Ionizing Radiation Detectors

Verification of electret properties, using TSQ and TSDC investigations, confirms that by selecting appropriate process parameters, such as temperature and charge implantation time, the corona charge method guarantees fabrication of PTFE foil electrets with the desired properties. In Figure 7,

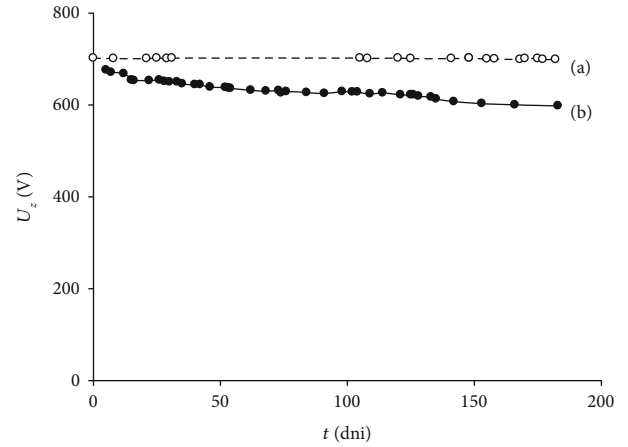


FIGURE 7: Time dependency of the electret equivalent voltage for a) a sample formed at an elevated temperature and b) a sample formed at ambient temperature.

the plot of the changes in the electret equivalent voltage as a function of time is shown for samples formed at ambient and elevated temperatures. The electrets optimal for use in the ionizing radiation sensors were fabricated under elevated temperature conditions. The process at ambient temperature does not guarantee a sufficiently long lifetime of the electrets [10].

Based on the obtained results of the sample equivalent voltage U_z , the trend of its changes in as a function of time was determined for electrets formed at ambient temperature and electrets formed at elevated temperatures. The results are shown in Figure 7. They show that charge of electrets formed at elevated temperature is more stable than the charge of electrets formed at ambient temperature [12].

3.1. The Construction of an Electret Dosimeter. A prototype of an electret dosimeter consists of an ionization chamber, shown in Figure 8, and an electromechanical unit, shown in Figure 9, for the measurement of the equivalent voltage. The electret (3), in the form of a flat disc made from a PTFE foil, is located at the bottom of the chamber. Using a corona charge system in an air triode configuration, the electret has been charged to a predetermined value of an uncompensated positive electric charge density (equivalent voltage). The measurement of radon concentration in the air of the investigated space begins when the hole cover is opened and the gas enters into the chamber through a filter preventing the decay products from entering the chamber. The half-life time of radon is 3.8 days and the radiation resulting from its decay chain ionizes the air inside the chamber. The negatively charged ions, attracted to the electret, release electrons causing compensation of the electret's positive charge. The measurement ends when the dosimeter chamber is closed [11].

Using an electrometric voltmeter, permanently attached at the top of the ionization chamber, the electret's equivalent voltage is measured before opening and after closing the ionization chamber. The measurement of the electret equivalent voltage is carried out by measuring the strength of the electric field E in the gap between the electret and the reference electrode of the voltmeter. Figure 9 shows the principle of the electric field measurement using an electric field chopper.

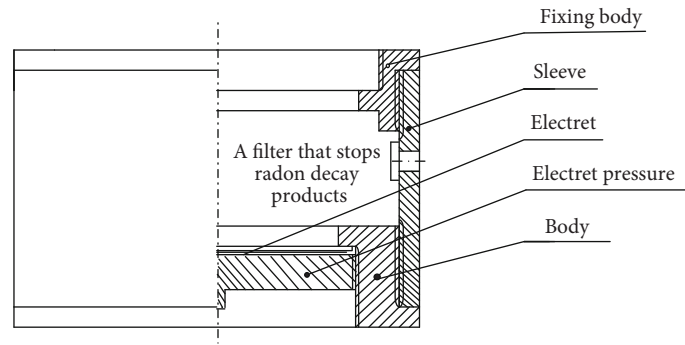


FIGURE 8: Ionization chamber of an electret dosimeter.

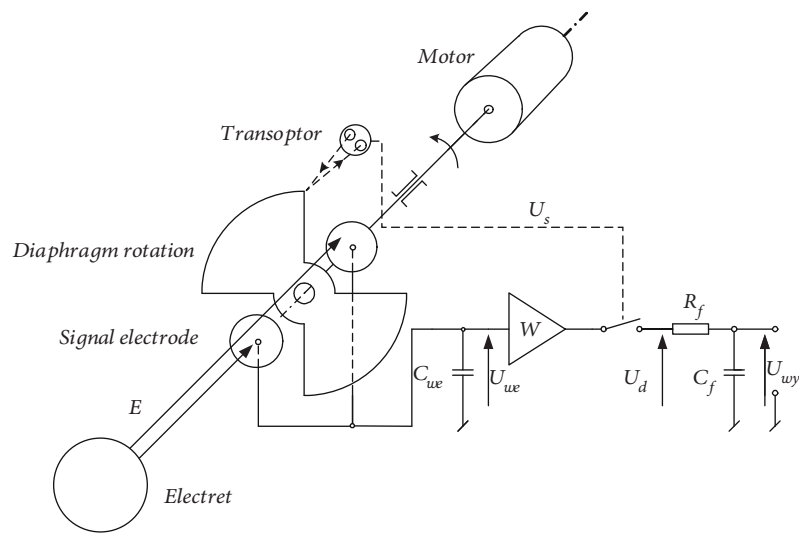


FIGURE 9: The measurement of the electric field using a field chopper.

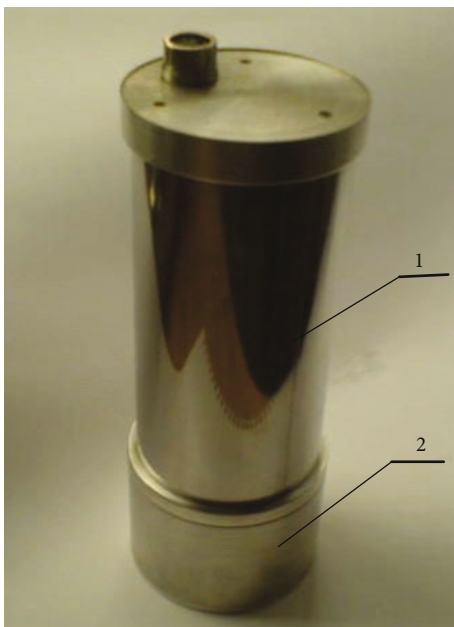


FIGURE 10: View of the electret dosimeter, 1 - electric field measuring system, 2 - ionization chamber with electret.

The chopper blades rotating in front of the reference electrode cyclically interrupt the electric field lines between the electret and the reference electrode. As a result, a constant electric field is converted into an alternating signal, proportional to the field intensity. The electric field meter is calibrated to directly display the value of the electret equivalent voltage [16].

In the solution here presented, the electrometric meter is mechanically attached to the ionization chamber with the electret. This design makes it possible to directly determine the value of the electret's equivalent voltage without the necessity of removing the electret from the chamber.

The complete dosimeter with an electret sensor is shown in Figure 10. It contains an ionization chamber with an electret, and a meter of the effective surface charge density of the electret, mechanically connected to the ionization chamber. The combination of the ionization chamber with the electric field measurement system is a novel prototype of an electret dosimeter.

3.2. Electret Testing in an Emanation Chamber. The formed electrets were investigated by exposure to calibrated radon irradiation. The process took place in a 50 l emanation chamber made by the GENITRON INSTRUMENTS GmbH. The chamber was filled with air containing radon from a leak

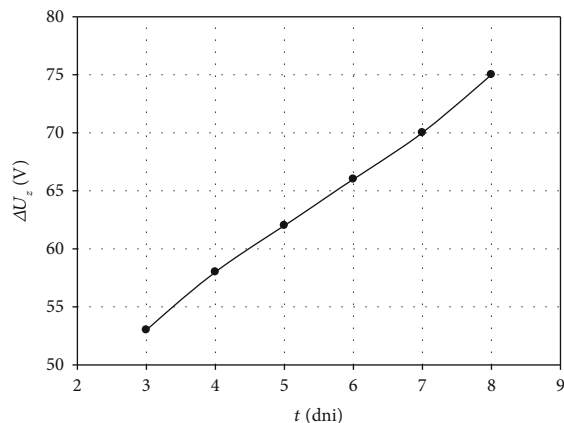


FIGURE 11: The change of the electret equivalent voltage from its initial value of 700 V versus the exposure time to radon radiation.

regulated SIST SRM- 4968 source with 50 Bq activity. Several multi-day (4-7 days) exposure cycles were separated by thirty-day breaks in order to achieve the required radon accumulation at a desired equilibrium level. From the obtained data, it was found that the average radon activity in the ionization chamber was 285.9 Bq and the average radon concentration in the chamber was 5648 Bq/m³. The change in the electret equivalent voltage as a function of the exposure time is shown in Figure 11.

The obtained plot clearly shows a linear dependence of the change in the effective surface charge density (equivalent voltage U_z) of the electret as a function of exposure time to the radiation resulting from radon decay.

4. Conclusions

The electrostatic tests carried out on electrets formed with the corona discharge method in the air triode configuration indicate that the method and processing conditions give repeatable values of the surface charge density (equivalent voltage U_z) of the obtained electrets.

The charge (equivalent voltage U_z) of electrets formed at an elevated temperature is more stable than that of electrets processed at ambient temperature.

Electrets produced by the corona discharge method at elevated temperature from the foils selected by the authors, have a sufficiently long life and can be used as elements in electret dosimeters for measurements of radon concentration.

This research suggests a linear relationship between the effective density of electret surface charge (equivalent voltage U_z) as a function of exposure time to ionizing radiation.

The average rate of change in the equivalent voltage of the electret in the dosimeter subjected to the effects of alpha particles produced in the ²²²Rn decay at an average radon concentration inside the test chamber of 5648 Bq/m³ was 4.4 V/day.

In order to obtain the highest possible stability of the electret equivalent voltage U_z required in the radon concentration measurements during its storage, it is necessary to perform aging investigations on the formed electrets in selected environmental conditions before the proper measurements.

The obtained results would allow optimization of the electret formation conditions in order to obtain an electret that is sufficiently sensitive to ionizing radiation.

Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

Conflicts of Interest

The authors declare that there is no conflict of interest regarding the publication of this paper.

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