Chapter 2

Radon Nuclides and Radon Generators

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Additional information is available at the end of the chapter

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Abstract

The radon element is the heaviest and the only naturally occurring radioactive noble gas. As a member of uranium and thorium decay chains, it is formed instantaneously and belongs to the naturally occurring radioactive materials (*NORM*). The long-lived radon isotope, the ^{222}Rn, is radiobiologically the most important one. It is present in subsoil and groundwater and permeates to the surface, where it may become health risk during the long-term inhalation. Proper testing of drinking water and building materials is also required to monitor radon concentrations below legal limits. Thus, the need of radon determination as well as the preparation of its isotopes arises for its use as a calibration source for the environmental and workplace monitoring in the *NORM* as well as other industries. Further, the radon isotopes currently appear in various research fields, including radionuclide progeny preparation and their use is experiencing renaissance. An overview of radon characteristics, its physical and chemical properties, as well as radon isotope preparation methods including the radionuclide generators and their use is given here. Radon isotope use for tracing, medical, geochemical and other purposes is also discussed.

Keywords: radon, thoron, actinon, generator, decay chain, radioactive deposits, Rn, noble gas

1. Historical introduction

Soon after the discovery of radium by Curie et al. [1], the radioactivity of thorium by Schmidt [2] and the discovery of actinium by Debierne [3], it was found that all of these radioactive elements activate their surroundings and emit formerly unknown radioactive gases. Curie and Curie discovered that objects exposed to radium samples got activated and the half-life of the gained radioactivity was approximately 1 month [4], even though clear interpretation of this discovery remained unknown. Similarly, radioactive gas evolution was observed

in samples of thorium and uranium [5–9]. Rutherford and Dorn interpreted the emanated activity from thorium and radium compounds as new chemical substances. Rutherford and Owens reported that the gas released from thorium had the half-life of about 1 min. Radioactive gas release from actinium samples was reported by Debierne soon after [10]. All the three gases (originating from radium, thorium and actinium) were found as chemically inert and were classified as noble gases [11]. Further studies on emanation gases lead to better understanding of their physical and chemical properties as well as their nature, for example, their condensation at low temperatures [12]. Many experiments were performed in order to determine the atomic mass of the emanation. In 1910, among others, Ramsay and Gray collected radium emanation gas to measure its density and determined the value of atomic mass to be around the value of 220 [13]. Later on, the formerly known and so-called radium, thorium and actinium emanations were named as radon, thoron and actinon, respectively [14]. The radon nuclides were originally important subjects of basic science [15]. After the determination and precise measurements of all their physical and chemical properties, various applications of radon appeared such as its use in medicine, geology, tracing applications, radionuclide production and research.

2. Radon nuclides and their characteristics

Radon (the element) is the heaviest known noble gas. Radon has only radioactive nuclides. In total, some 40 isotopes of radon are known up today. Three most common and naturally abundant isotopes of radon are ^{222}Rn, ^{220}Rn and ^{219}Rn, originating from the decay series of ^{238}U, ^{232}Th and ^{235}U, respectively. Their decay chains are shown in **Figures 1–3**, and the nuclear data are summarized in **Table 1**. Nuclear data were taken from Refs. [16, 17].

Some selected physical and chemical properties of radon element are summarized in **Table 2**. Data were taken from Ref. [18]. Radon belongs to the noble gas group that predicts its parameters.

As it could be clearly seen from **Figures 1–3**, the daughter nuclides of radon generate radioactive progeny of Po, At, Pb, Bi, Hg and Tl. The decay of Radon in air (gas phase) results in the spread of radioactive aerosols and their deposits mainly in case of ^{222}Rn may become hazardous (long-lived deposits). Radiotoxicity of radon is thus an important factor, since it may be released from natural sources (e.g., building materials, subsoil, drinking water and mine air) and inhaled.

Following the natural decay series (^{238}U, ^{232}Th and ^{235}U), the nuclides of ^{222}Rn, ^{220}Rn and ^{219}Rn are instantaneously formed progeny in the environment and their mother nuclei samples. These isotopes may be, however, produced in various nuclear reactions, for example, the ^{219}Rn was co-produced in the experiment of chemical characterization of the element copernicium (^{283}112) in the nuclear fusion of ^{48}Ca with ^{242}Pu, where the ^{219}Rn served as a calibration nuclide in the thermochromatographic separations of the element 112 in the *COLD* detector [19]. Other exotic isotopes of radon may be produced in various nuclear reactions. For example, the lightest-known neutron deficient isotopes of radon, ^{193}Rn and ^{194}Rn, were produced in the complete fusion reaction of ^{52}Cr pulsed ion beam with a ^{144}Sm target [20]. The ^{209}Rn and ^{210}Rn isotopes were determined in the decay chains of ^{213}Ra and ^{214}Ra, respectively, prepared

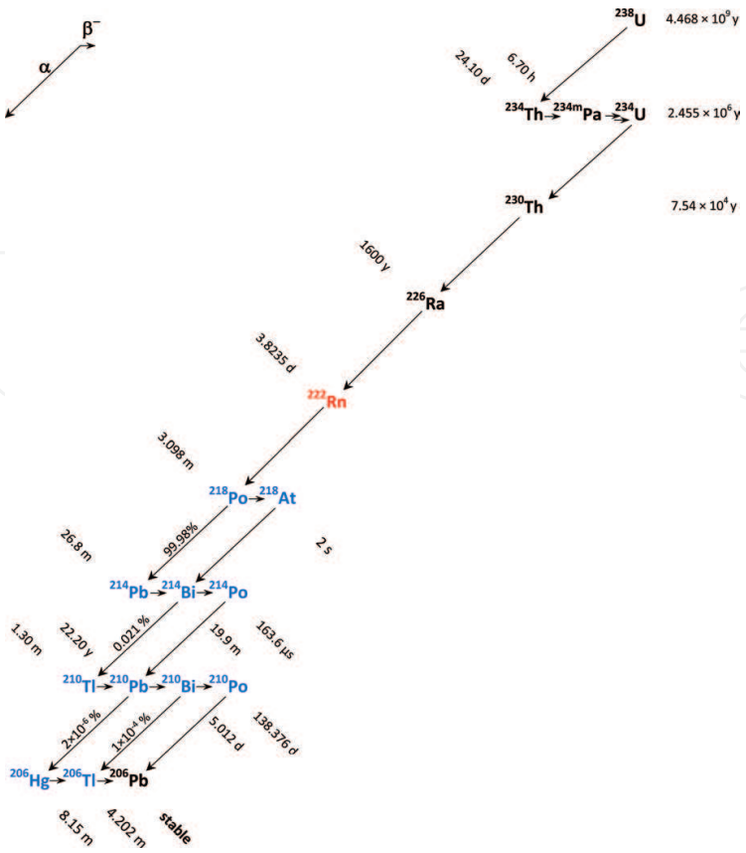


Figure 1. ^{238}U decay chain.

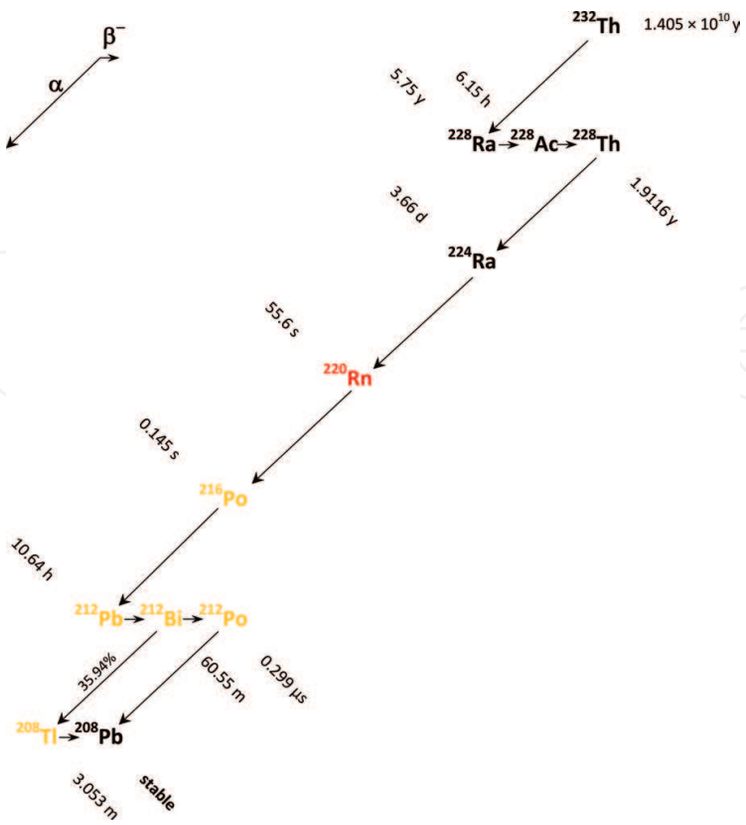


Figure 2. ^{235}U decay chain.

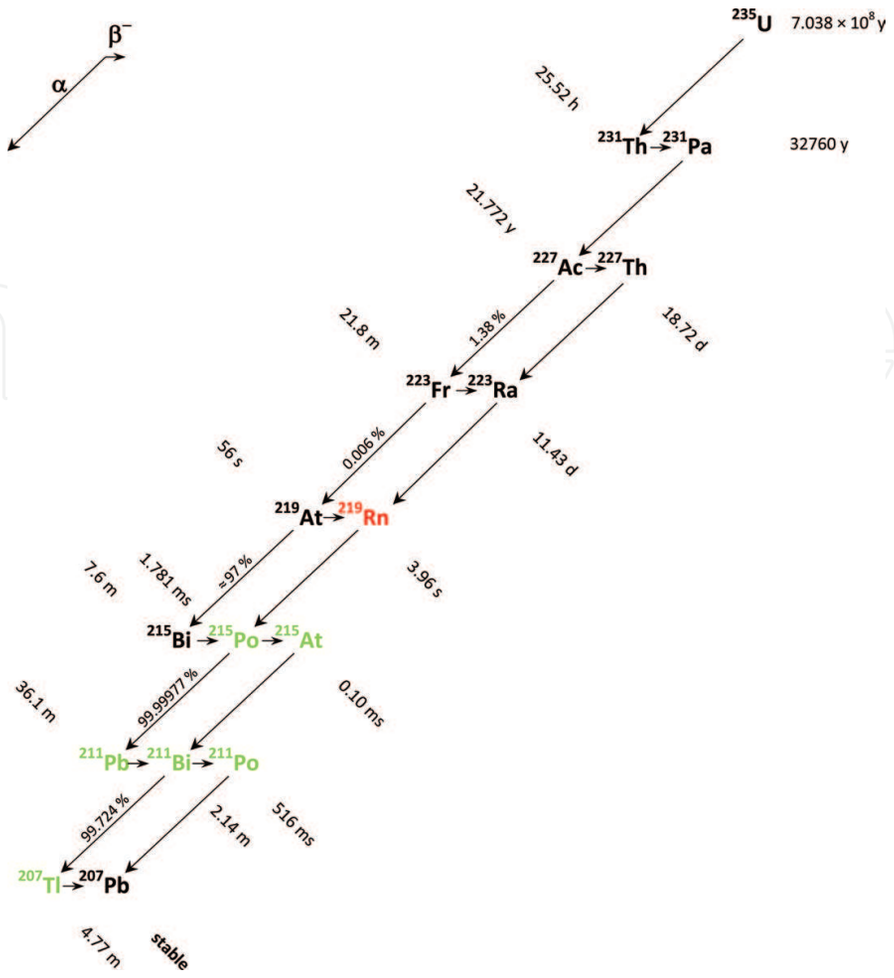


Figure 3. ^{232}Th decay chain.

Historical name	Radon	Thoron	Actinon
Nuclide	^{222}Rn	^{220}Rn	^{219}Rn
Half-life	3.8253 d	55.6 s	3.96 s
Generator nuclide	^{226}Ra (^{238}U) 1600 y	^{232}Th (^{228}Th) 1.405×10^{10} y	^{227}Ac (^{235}U/^{231}Pa) 21.772 y
Main α radiations E (MeV) (intensity (%))	5.4895 (99.92) 4.987 (0.078) 4.827 (~0.0005)	6.288 (99.886) 5.747 (0.114)	6.819 (79.4) 6.553 (12.9) 6.425 (7.5) 6.529 (0.12) 6.312 (0.054) 6.159 (0.0174)
Main γ radiations E (keV) (intensity (%))	511 (0.076)	549.8 (0.114)	271.2 (10.8) 401.8 (6.37) 130.6 (0.119) 293.5 (0.073) 517.6 (0.0443) 221.5 (0.030)

Table 1. Main isotopes of radon.

Electron shell configuration	[Xe] $4f^{14}$ $5d^{10}$ $6s^2$ $6p^6$	
Oxidation numbers	2, 3, 4	
Ionization potential (eV)	1st	10.748
	2nd	21.4
	3rd	29.4
	4th	44
Pauling electronegativity	2.0	
Atomic radius (nm)	218	
Molar enthalpy of fusion at (kJ mol^{-1})	12.26	
Molar enthalpy of vaporization (kJ mol^{-1})	74.30	
Surface energy (mJ m^{-2})	29	
Melting point (°C)	−71	
Boiling point (°C)	−61.8	
Solubility of radon at standard pressure in water (cm^3/100 g H_2O)	0°C	51.0
	50°C	13.0
Density (g cm^{-3})	0°C (g)	0.00973
	−61.8°C (l)	4.4
Dynamic viscosity coefficient (mPa s)	0.0213	
Thermal conductivity coefficient at 27°C (W m^{-1} K^{-1})	0.00364	

Table 2. Selected physical and chemical properties of radon.

via $^{170}Er(^{50}Ti,3n)^{217}Th \rightarrow {}^{213}Ra$ and $^{170}Er(^{48}Ca,xn)^{218\text{-}x}Ra$ reactions [21]. The ^{211}Rn was prepared as a decay product of the francium beams produced by the spallation of actinide targets (U and Th) [22]. The ^{211}Rn was also prepared by the $^{209}Bi(^{7}Li,5n)^{211}Rn$ reaction [23]. Heavier isotopes such as ^{224}Rn, ^{223}Rn and lighter isotope of ^{210}Rn were prepared by the spallation of $^{232}ThO_2$ [24, 25]. The neutron-rich isotopes of ^{227}Rn and ^{228}Rn were also prepared by the spallation of natural ^{232}Th target with 600 MeV proton beam [26]. The heaviest experimentally detected isotopes of radon, the $^{223–229}Rn$, have been determined for the first time, using the *ISOLTRAP* setup at *CERN ISOLDE* experiment [27].

Radon is chemically quite unreactive gas; however, some exotic compounds of radon, for example, the fluorine compounds of radon [28] or RnH^+, $RnOH^+$, $RnOH_2^+$ molecular ions generated in a plasma ion source [29] were reported; other compounds like RnCO [30], HRnCCH [31] and other radon molecules [32] were predicted. Radon adsorption on charcoal is known for a long time and allows its purification from hydrogen, oxygen and nitrogen [33]. Recently, the association of xenon and radon with tris-(triazole ethylamine) cryptophane was studied. High affinity of these noble gases was observed, and the association constants were determined to K_a = 42,000 ± 2000 M^{-1} and K_a = 49,000 ± 12,000 M^{-1} for xenon and radon, respectively, at 293 K [34].

3. Radon generators

Thanks to the noble gas characteristics of the radon element, its separation from the decay chain or target materials becomes trivial. Radon generators may be then divided into two main groups based on the medium that is used for its final form—gas and liquid apparatuses. Main issue in radon generators is the emanation efficiency, radon physical form and its radionuclidic purity. The radon emanation power or the efficiency coefficient depends directly on the radon source properties. Crystalline and bulky materials exhibit quite low emanation power—typically few per cent of the mother nuclide activity. On the other hand, the emanation efficiency of properly selected and prepared amorphous and porous materials approaches the values of nearly 100%. The simplest generators include simply the mother nuclides enclosed in an evacuated or normal pressure apparatus, allowing the liberation of radon that is to be collected in a gas or liquid phase. Typically, the insoluble salts of mother nuclides trapped in a porous ceramics (or other inert material) or a sandwich of mother nuclide covered by thin separating layer (e.g., foil) were used for the construction of various types of emanators.

Many radium-based inhalation apparatuses as well as drinking or bath water "activators" (**Figure 4**) with the emanation power in the order of 5000–10,000 *Mache* units in 24 h appeared on marker without any regulation (1 *Mache* unit = 3.64 *Eman* = 3.64 × 10^{-10} Ci/L = 13.4545 Bq/L).

Further, more advanced systems were developed in order to increase the efficiency and purity of the radon gas. For example, the oxygen and ozone may be chemically removed by passing the radon gas through the heated copper wire; the hydrogen by the heated copper

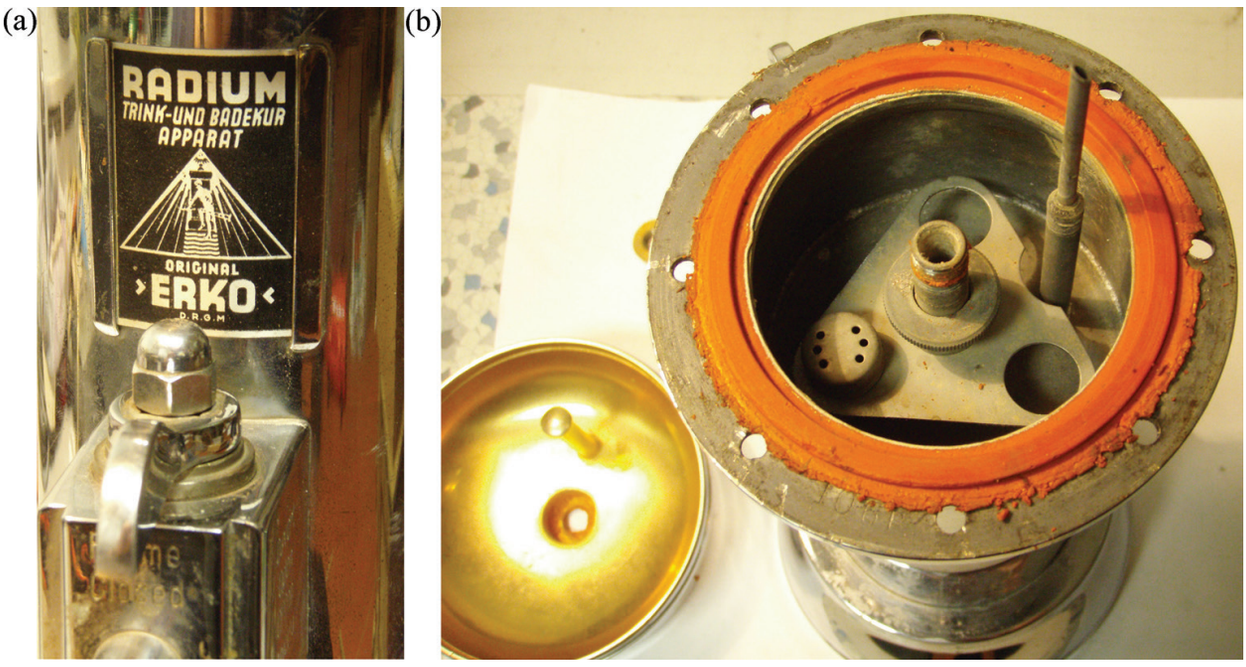


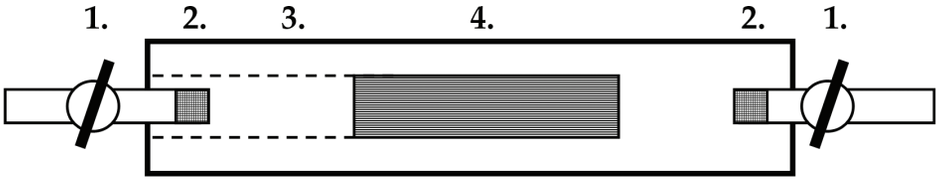
Figure 4. Left—the "ERKO" instrument for the preparation of radium water produced by the Berliner Radium Aktiengesellschaft in 1930s. Right—the same instrument in detail showing a carousel holding one radium capsule with holes to allow free radon emanation (air/water).

oxide; CO_2 by its capture on potassium hydroxide and finally the water on phosphorus pentoxide. Radon of specific activity of 18.5 GBq/mm^3 could be prepared in such way (approx one-third of theoretical volume activity value of pure ^{222}Rn) [35]. Various materials with high emanation power were developed and tested for the construction of radon generators, for example, inorganic porous gels based on heavy and alkali-earth metal hydroxides with hydrated silicic acid were developed [36, 37]. Also the barium stearate powder was reported with the emanating power of >99% for thin layers in air at atmospheric or reduced pressures for actinon [38].

Another important aspect is the source activity metrological standardization. This is necessary firstly for the precise determination of radon contents in various materials and secondly to test the material permeability or retention ability for radon and to verify that these materials meet legal regulations. Various methods for the preparation of calibrated emanation standards were published. Standards containing the solution of $^{226}Ra(NO_3)_2$ absorbed into $CaCO_3$ were prepared, and the emanation coefficient of ^{222}Rn for these standards varied from 0.23 to 0.25 [39]. Accurate and long-term stable sources of defined activity of ^{222}Rn in gas phase were developed for laboratory and field applications (**Figure 5**) [40]. Radon is released from thin layer of a plastic foil with emanation power coefficient approaching 1. The source is constructed as a stainless steel cylinder supplied with the two ball valves on the ends and the two aerosol filters connected on the output aperture of the valves.

Several systems for the preparation of radon in water standard sources were reported. Standard based on an earlier and previously described prototype consisting of polyethylene-encapsulated ^{226}Ra solution source in a small-volume accumulation chamber was used to generate and accurately dispense radium-free ^{222}Rn solutions of known concentration [41]. More recent radon in water standard was developed to get the radon solutions of 300–2000 Bq/L [42] The generator consisted of about 6 L cylindrical vessel with a solid phase ^{222}Rn source with 99.9% air emanation power and an external circuit for solution homogenization. Another radon generator and delivery system was used with 2.9 GBq of radium salt for cell cultures exposure studies [43].

Interesting technique for the radon source preparation that is suitable for use in a low-background liquid scintillation detectors was reported [44]. Radon was concentrated from air to prepare liquid scintillation counting (LSC) sources spiked with activities of 10^6 Bq/m^3. CO_2 and water vapour were removed, and the radon was collected in a cooled charcoal trap. The



1 – Ball valve, 2 – aerosol filter, 3 – holder, 4 – emanator with ^{226}Ra

Figure 5. Drawing of radon flow-through generator (air/air).

accumulated radon was desorbed and transferred into a 1,2,4-trimethylbenzene-based scintillator. The sources have been used for the calibration.

Simple laboratory demonstration apparatus for thoron (^{220}Rn) preparation could be constructed, using aged sample of thorium nitrate solution [45]. The solution is enclosed in a bubbler flask connected to a small compressor from where the flowing air displaces the thoron through the valves. Lucas-type scintillation counter [46], ionization chamber or a cloud chamber could be directly connected to the emanation flask through the drying column. Thoron half-life could be easily determined by counting the gas activity enclosed in the detector over few minutes or alternatively the alpha-particle tracks could be visualized in a cloud chamber (**Figure 6**) for educational and demonstration purposes.

Figure 6. Tracks of alpha particles from thoron (^{220}Rn) decay visible in an isopropanol-filled continuous cloud chamber.

4. Radon counting

Measurement of emanation ionization-induced discharge of an electroscope was the first method to determine its activity. Later on, ionization, scintillation and many other types of detectors were developed for radon counting. Various passive detectors as well as flow-through detectors were developed. Also the direct radon detection techniques, radon progeny detection and even electronic nuclear track detectors were developed.

The direct and precise radon activity determination (typically volume activity in air or water) is, due to radon physical and chemical properties, problematic since the inert gas may escape the sample and the volume activity depends on many factors (e.g., the weather and building ventilation). Thus, particular precautions need to be applied for sample treatment. For example, the well water must be stored in tight bottles in a cold place and measured within few days from the sample collection.

Short-term radon monitoring in air could be easily performed by the measurement of defined amount of air enclosed in a Lucas cell-type detectors, based on ZnS(Ag) scintillation material [46]. Various geometries were developed, and even 1 L cell volume detectors are available with minimal significant volume activities in the range of 2 and 3 mBq/L for airborne radon and ^{222}Rn in water, respectively [47]. Measurement of ^{222}Rn was also performed by its absorption in a plastic scintillators and alpha/beta pulse shape discrimination [48]. These techniques may be used in radon risk determination on a building sites. Soil air is taken from the ground by drilling several exploratory wells up to the depth of 1 m, and air samples are collected with air-tight syringe, transferred to a Lucas cell and counted. Further, the radon gas permeability through the soil and fundaments is also evaluated to determine overall risk [49]. Even though certified methods were applied more detailed analysis is needed in some cases to provide accurate results [50]. For some systems and low-background radon counting, the decayed air or low radon gas is needed to reach low detection limits. Such apparatus for low radon nitrogen was reported [51].

Another type of detectors for longer determination periods (e.g., 2 weeks) are the electret dosimeters [52]. Integral measurement gives an average value of radon volume activity in the air and is less influenced by temporary short-term changes in the monitored place situation.

Long-term determinations are performed with radon nuclear track detectors [53]. These are placed in a monitored area and left for the period of even several years to collect the tracks. These are further etched and detected under microscope and evaluated using CCD camera and PC software [54]. The comparison of various types of alpha-track detectors was evaluated some time ago [55].

For laboratory measurements, semiconductor detectors for alpha and gamma spectroscopy may be used with an advantage of radon, thoron and their progenies discrimination; however, the need of enclosed apparatus is crucial in direct measurements and usually allows only the determination of the progeny. Interesting and a very simple emanation method for determining radium was described, where the radon was adsorbed on a silica gel at the temperature of liquid nitrogen and then transferred at 0°C to a toluene-based liquid scintillator [56].

Other techniques for simultaneous measurement of radon and thoron were studied, for example, using the Timepix electronic nuclear track detector [57]. Electronic radon/thoron detection system was developed, employing the passivated ion-implanted planar silicon (PIPS) detector [58].

A method that allows to distinguish surface radon sources from the deep sources was reported recently [59]. The method for the determination of the relative depth of a radon source is based on the field alpha-spectroscopy of radon (^{222}Rn) and actinon (^{219}Rn) progenies in soil gas. The limitations of the determination were obvious, firstly the 40 s time-period (half-life of actinon) and secondly the geological-structural situation of the studied locality where the high value of the activity ratio of ^{211}Bi and ^{214}Po corresponded to a situation where the short-lived isotope ^{219}Rn was present in the sample in larger amounts than that corresponding to the natural ratio.

Personal monitoring of radiation workers, mainly in uranium industry and other mines in uranium-rich regions, includes a combination of several types of detectors in order to properly estimate the acquired cumulative dose and to discriminate the inhaled radiation burden. Thus they typically contain a thermoluminescent detector for external gamma radiation

dose measurement, active flow-through filter unit for the measurement of long-lived progeny aerosols and a nuclear track detector typically equipped with energy-absorbing foils for the radon and thoron alpha decay discrimination [60].

5. Applications and use of radon

The radon element applications include various research, industrial and other fields like its use in spa-based therapies and the exploitation of its natural occurrence in anomalous quantities.

From the early times, radon emanation and the emanation method were applied in the studies of material structure, for example, to determine specific surface, material porosity and their crystalline/amorphous structure [61]. Recently, novel method and an installation for rapid determination of the radon diffusion coefficients in various materials were described recently [62]. Such measurement is important for the development and characterization of radon barriers. Emanation method was also used for the estimation of reactivity of ferric oxides prepared from different sources [63].

To study the radon isotope permeation through the barriers as well as to perform dosimetric studies and other experiments under radon exposure, radon chambers are constructed. A review of different radon chambers with volumes from 0.01 up to 78 m^3 appeared, describing several setups [64]. Fully automated radon chamber was also developed, including the controlled atmosphere (humidity, pressure and radon activity) [65]. The chamber of 1.46 m^3 was made of stainless steel and allowed ^{222}Rn, ^{220}Rn or both to be injected from the bottom pipelines into the chamber in a 100% flow-through mode, 100% recirculate mode or flow-through/recirculate mode.

Another application of radon includes the labelling of surface layers using the active deposits of radon. That is possible, thanks to the nuclear recoil effect and their electrostatic deposition. This method allows to prepare, for example, surface-labelled solid samples and to perform their wear tests [66].

Since the radon gas permeates the underground through the rock cracks and enters into springs and soil air, it may be useful for uranium and other ores prospecting by the emanation detection of radiometric anomalies [67]. Interesting fact of exterrestrial radon occurrence was reported on the lunar surface [68]. Very important geophysical application of radon detection is the measurement of radon release anomalies preceding the earthquakes [69].

Another isotope of radon, the thoron, was used in the separation of ^{212}Pb from ^{228}Th, allowing the construction of a radionuclide generator [70] and its use for the labelling of radioimmunoconjugates for targeted alpha-particle therapy [71]. The production of ^{212}Pb is nowadays performed under good manufacturing practice (GMP), and clinical trials of several tracers are ongoing e.g. [72].

The use of radon (^{222}Rn) in medicine and spas radically decreased as soon as the deterministic effects of ionizing radiation on humans were better understood. On the other hand, under proper regulation, low-activity radon therapy is even nowadays beneficial for the patients suffering from painful inflammatory rheumatic diseases, diseases of musculoskeletal system, diseases

of the peripheral nervous system, diabetes and others. In Jáchymov, Czech Republic, the world-famous radium spa is operating four water springs with maximal radon content of 5–20 kBq/L [73]. Despite of insufficient number of clinical trials [74] and some controversies in the radon therapies [75, 76], the long-term experience demonstrably confirms the benefits for the patients and justifies its use, further confirmed by many independent scientific studies [77–80]. The spa medical praxis was verified by decades and is supported also by other epidemiologic studies on the hormesis theory that supports the beneficial effects of low-dose radon exposure [81–83].

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