

INDOOR RADON MEASUREMENTS BY NUCLEAR TRACK DETECTORS: APPLICATIONS IN SECONDARY SCHOOLS

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Abstract. *Indoor radon measurements by nuclear track detectors and application of the method in secondary schools in Serbia were performed in the spring 2004. Thirty detectors (type CR-39) were distributed to high school teachers in several cities in Serbia. After three months of the detectors exposure, they were sent back to the Low-Level Laboratory, Institute of Physics, Belgrade. After exposure, the CR-39 detectors were etched in a 6N NaOH at 70°C for 3 hours. The tracks were counted by the semi-automatic track-counting system. The preliminary results are presented in this paper.*

Key words: *Radon, nuclear track detectors*

INTRODUCTION

Radon is a unique natural element in being a gas, noble, and radioactive in all of its isotopes. As gases the isotopes are mobile and carry messages over significant distances within the earth and in the atmosphere, but, on the other side, inhalation can be a problem to health. The fact that radon is noble ensures that it is not immobilized by chemically reacting with the medium that it permeates. The only way that radon diminishes is radioactive decay. Its radioactivity allows radon to be measured with high sensitivity. Unfortunately, high radon concentration can be a health risk, a cause of lung cancer. The detection and radon concentration measurements are one of the most important procedures in environmental protection.

The passage of heavily ionizing, nuclear particles through majority of insulating solids creates narrow paths of intense damage to an atomic scale. These latent tracks are distinguishable from the intrinsic irregularity and damages to the crystal structure. In addition, they may be revealed and become visible under an ordinary optical microscope when treated with a properly chosen chemical reagent. This effect was discovered on LiF crystal in the late fifties of the last century. Held in contact with a uranium foil and exposed to thermal neutrons, LiF crystal revealed a number of etch pits after treatment with

a chemical reagent. The number of these pits showed complete correspondence with the estimated number of fission fragments which would have recoiled into the crystal from the uranium foil [1], [2]. Soon after discovery of the first track detector, many other crystals were found with the same properties. Besides fission fragments tracks, it is also possible to reveal tracks of other heavy ions. The next big step in the developing of this new investigation area was the discovery that some kind of plastic materials are nuclear track detectors and that some of them are able to detect alpha particles and protons. A few theoretical models for track formation mechanism, track etching methodology and geometry are described in the first part of this paper. Everything about principles, methods and application of track detection in solids can be found in reference books [3] and [4].

Because of their simplicity, good geometry and possibility to get an integral signal after long-term measurements (up to several years), nuclear track detectors are widely used in different scientific disciplines. Their use ranges from fundamental investigations (search for super-heavy elements, magnet monopole, etc.) through high and low energy nuclear physics, geology to dosimetry and radon measurements. Radon measurements by etched track detectors are the goal of this paper. Passive radon monitoring devices based on alpha particle etched track detectors are very attractive for the assessment of long-term radon exposures, especially for radon mapping. Radon mapping is performed for identifying locations of permanent features of the Earth's crust or of its contents. In this case, radon mapping over a long time (at least one climatic cycle) is needed because of seasonal and nyctemeral variations of the radon response to environmental stress. Up to now, there has been no serious attempt to get a radon map of the Serbian territory (except for the radon mapping of Vojvodina [5]). In that sense, thirty nuclear track detectors (type CR-39) were distributed to high school teachers in several cities in Serbia. After three months of detectors exposure, they were sent back to the Low-Level Laboratory, Institute of Physics, Belgrade. After exposure, the CR-39 detectors were etched in a 6N NaOH at 70°C for 3 hours. The tracks were counted by the semi-automatic track-counting system. The preliminary results are presented in this paper.

TRACK FORMATION MECHANISMS, TRACK ETCHING METHODOLOGY AND GEOMETRY

Etched tracks have now been observed in a large number of materials. These materials are generally polymers, inorganic glasses, mineral crystals and some poor semiconductors. The most important generalization that can be made about these substances is that they are all dielectric solids. Free electrons in metals destroy a latent track formed by incident ion in the recombination process with the ion-holes of the media before etching treatment. The limiting resistance below which tracks are not observed is empirically determined to be 2000 Ωcm . Other criteria and theoretical models for track formation mechanisms have been closer to the physical reality based on interactions of charged particles with the matter.

Energy losses of the incident ions are the result of interactions with electrons and nuclei of the media. For ions with energies greater than 1 MeV, nuclear losses are small compared to electronic energy losses. On the other side, heavy ions dominantly lose energy primarily through Coulomb interactions with orbital electrons of the target atoms. These interactions are described in the Bethe-Bloch formula:

$$\left(\frac{dE}{dx}\right) = \frac{C_1 Z^2}{\beta^2} \left[\ln\left(\frac{W_{\max}^2}{I^2}\right) - 2\beta^2 - \delta - U \right] \quad (1)$$

Here, $C_1 = 2\pi n_e e^4 / mc^2$, n_e – electron concentration, m – electron mass, W_{\max} – maximum of the energy transfer, $\beta = v/c$, v – ion velocity, I – mean ionization potential, δ – correction on the polarization effect of the media for relativistic velocity, U – correction for small velocity.

The earliest explanation of track formation was that it depends on a total amount of energy deposited per unit path length by the incident ion. Total energy losses (dE/dx) in interactions with electrons are the function of ion energy and have a characteristic shape (Figure 1). This shape is directly related to the empirically observed critical value of $(dE/dx)_c$. When (dE/dx) exceeds this critical value, the tracks are formed. This approach became untenable in the high-energy region. The tracks are not revealed because high-energy ions produce a large number of δ electrons. These electrons take off most of the accepted energy out of the track area so it is lost in the etching process. For that reason various authors have suggested that track formation should be related to a number of different parameters (some of them are shown in Figure 1).

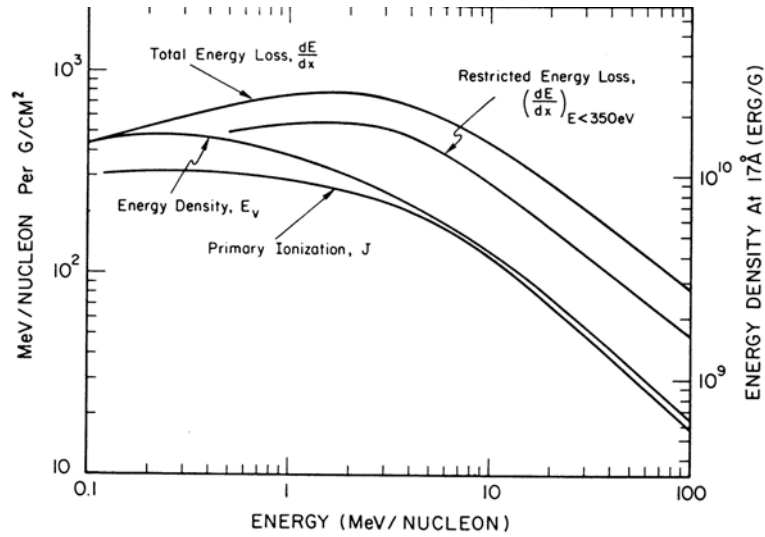


Fig. 1. Various track formation criteria as the function of heavy ions energy.

One of these criteria is the number of primary ionizations produced close to the ion path. The relation for primary ionization J , based on the Bethe-Bloch formula is:

$$J = \frac{C_1 C_2 Z^{*2}}{I_0 \beta^2} \left[\ln \left(\frac{W_{\max}}{I_0} \right) - \beta^2 - \delta + K \right] \quad (2)$$

Figure 2 shows the function of primary ionization J for different heavy ions and ion energies and detection threshold for the given detectors.

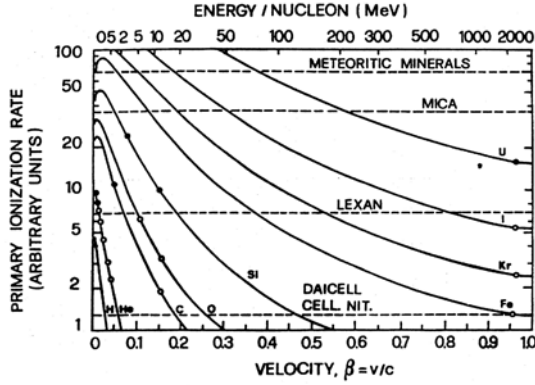


Fig. 2. Primary ionization as the function of heavy ions velocity (energy) for different detectors

Nuclear tracks formed by heavy ions are very small (only some tens of nm in diameter). These latent tracks can only be seen using an electronic (TEM, SEM, etc.) microscope. Fortunately, any of the nuclear track detectors has an appropriate etchant the basic characteristics of which are that the track etching velocity V_T along the trajectory of the ions is larger than the bulk etching velocity V_B for the etchant used under given etching conditions. The ratio V_T/V_B is an important parameter that is very useful in the identification of the charged particles in given detector media. This

ratio also gives us parameters such as "etching efficiency" and the critical angle of etching θ_c . The meaning of the above-defined quantities is explained in Figure 3, which is a usual representation of track geometry. The left side of Figure 3 shows the track etching geometry for the ions, perpendicularly incident to the detector surface.

When the incident angle of the ions is lower than 90° , the component of V_T perpendicular to the surface, $V_T \sin \theta$, must exceed V_B in order to produce a track (right side of Fig. 3). Hence, there exists a critical angle of the registration given by $\theta_c = \arcsin (V_B/V_T)$, below which the damage trail is not developed into a track.

The track etching velocity V_T along the trajectory of the charged particle in the given detector media is the function of the charged particle type and energy. For high Z (fission fragments) the ratio V_T/V_B achieves a value of 10^4 so the tracks have a long, needle-like shape with a little hole compared with the track length (the diameter D after etching time t is $2V_B t$ and the ion range is $V_T t$). In the case of alpha particles, the ratio V_T/V_B is not so big (for CR-39 it is between 2 and 3). The quantities V_T and V_B are dependent on temperature and etchant concentration (typically, it is HF, H_3PO_4 , NaOH, KOH, etc.) Chemical etching is usually carried out in a thermostatically controlled bath at temperatures ranging from $40^\circ C$ to $70^\circ C$, and the etchant is in aqueous solution at a molarity from 2 to 6 M. Typical etching times range from 2 to 6 h. When these etching parameters increase, then the resulting etch pit becomes enlarged.

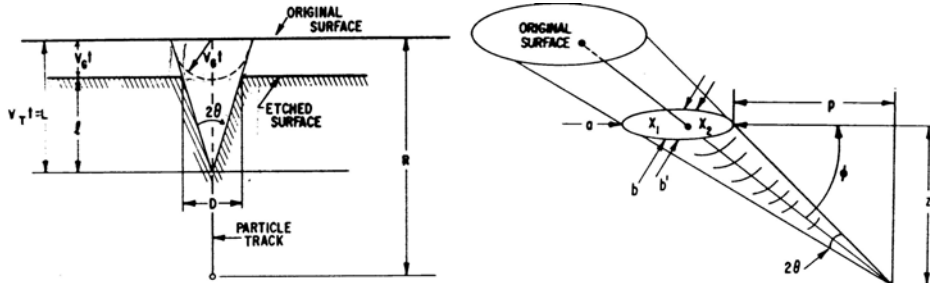


Fig. 3. Track geometry.

RADON MEASUREMENT BY NUCLEAR TRACK DETECTORS

The three radon isotopes are produced from radium decay as steps in lengthy sequences of decays that originate from natural radioactive series: ^{222}Rn (radon) from ^{238}U , ^{220}Rn (thoron) from ^{232}Th and ^{219}Rn (action) from ^{235}U . The relative importance of the isotopes increases with their half-lives and relative abundance. Action is the shortest lived ($\tau_{1/2} = 3.96$ s) and is practically always produced in much smaller amounts than is radon ($\tau_{1/2} = 3.82$ days), since the natural $^{235}\text{U}/^{238}\text{U}$ ratio is 0.00719. Thoron ($\tau_{1/2} = 55.6$ s) is short lived relative to radon and crosses over a much smaller distance from its source than does radon. Especially in the region where Th/U ratio is very high, the amounts of thoron are enhanced relative to radon (Kerala State in India, [8]). Radon and its isotopes emit alpha particles with different energies through radioactive decay. Energies and ranges of alpha particles in the air and detectors media are very important for detection and measurement of radon concentration. Table 1 shows the basic characteristics of radon, thoron and their progeny.

Radon measurements are based on its radioactivity. Radon is an alpha emitter, but some of its daughters are β and gamma radioactive. These properties allow numerous opportunities for the use of various detection and measurement techniques to get the information about radon concentration in the given media. Passive devices for radon measurements based on the nuclear track detectors are the subject of this paper. These types of the radon dosimeters are almost perfect for long-term measurements (radon mapping, for example). In this case, radon mapping over a long time (at least one climatic cycle) is needed because of the seasonal variations of radon concentration during one calendar year.

Table 1. Basic characteristics of the radon, thoron and their progeny

	Progeny	$\tau_{1/2}$	Alpha Energy (MeV)	Alpha Range in the air (cm)	Mean Diffusion Distance (cm)		
					Air	Porous Soil	Water
Radon	^{222}Rn	3.82 days	5.49	4.08	220	155	2.2
	^{218}Po	3 min.	6.00	4.67			
	^{214}Pb	26.8 min.	7.69	6.91			
Thoron	^{220}Rn	55.6 s	6.29	5.01	2.85	2.0	0.0285
	^{216}Po	0.15 s	6.78	5.67			
	^{212}Pb	10.6 h	8.78	8.53			
	^{212}Bi	60.55 min.	6.05	4.73			

Radon monitoring devices can be classified into four broad categories: (a) envelope or bare detector samplers; (b) diffusion samplers; (c) permeation samplers; and (d) radon collector samplers. Because of its good sensitivity, stability against various environmental factors and high degree of optical clarity, CR-39 has become the state-of-the-art etched track detector for environmental radon.

For dosimetric purposes, it is important to measure the average radon and its progeny concentration over a time that should be long enough relative to the typical time scale of radon fluctuations caused by environmental conditions. The signal measured by etched-track detectors is integrated track density, ρ (tracks cm^{-2}). If track density is uniform over the detector surface, the signal is defined by [6]:

$$\rho = \frac{K}{\tau_r} \int_0^{t_e} C(t) dt \quad (3)$$

where τ_r is radon decay time (5.5 days), $C(t)$ is radon concentration in the air around the detector (atoms m^{-3}) at time t , and t_e is the exposure time. K denotes the dosimeter response (calibration coefficient) and is defined by [7]:

$$K = \frac{\tau_r}{C(t)} \frac{d\rho}{dt} = \frac{1}{A_0(t)} \frac{d\rho}{dt} \quad (4)$$

where $A_0(t)$ is the activity concentration of radon in the air ($Bq\ m^{-3}$) at time t , and $d\rho/dt$ is the track density production rate (tracks $cm^{-2}\ h^{-1}$). For dosimeters with a long exposure time ($t_e > 3\ h$), the response can be approximated by [7]:

$$K = \frac{\rho}{\langle A_0 \rangle t_e} \quad (5)$$

where $\langle A_0 \rangle$ denotes the average radon activity concentration. On the other side, on a quite different approach [4], the expression in (6) is often employed for track density:

$$\rho_i = \frac{1}{4} A_i t_e R_i \cos^2 \theta_{c,i} \quad (6)$$

where R_i is the effective range of alpha particles emitted by the i -th nuclide in the air and $\cos^2 \theta_i$ is etching efficiency. Taking into account that θ in a CR-39 detector under normal chemical etching conditions is less than 20° and ranges of radon and its progeny alpha particles (Tab. 1) as well, from equations (5) and (6) we obtain a theoretical limit for the response of a bare detector, K to radon:

$$K = 3.2\ \text{tracks}\ cm^{-2}/kBq\ m^{-3}\ h^{-1} \quad (\text{CR-39}) \quad (7)$$

Everything about radon measurements by nuclear track detectors may be found in book [8].

EXPERIMENT

The radon problem is not taken into account seriously and systematically in our country. Recently published results [5] show that radio-ecological problems in residence buildings are not negligible even in the flat agricultural region such as Vojvodina. Almost 20% of radon concentration measurements are over $200\ Bq/m^{-3}$ and 4% of the all measurement places have significantly elevated indoor radon concentrations.

One of the possible solutions to that problem is given in this paper. Indoor radon measurements by nuclear track detectors and the application of the method in secondary schools in Serbia were performed in the spring of 2004. Thirty bare detectors (type CR-39) were distributed to high school teachers in several cities in Serbia. The measurement locations were different indoor places, from cellars to the garrets. After three months of exposure, the detectors were sent back to the Low-Level Laboratory, Institute of Physics, Belgrade. In our laboratory, there is a long and successful tradition of radon measurements with etched track detectors [9], [10].

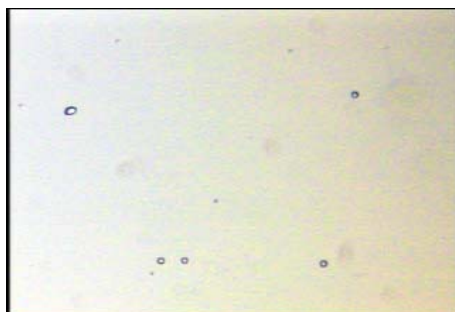


Fig. 4. Tracks of alpha particles emitted by radon in a CR-39 detector, which was exposed for three months in the region of Zaječar. One viewing field from the microscope has the area of about 0.5 mm^2 .

After exposure, the CR-39 detectors were etched in a 6N NaOH at 70°C for 3 hours. The etching time was shorter than usual because the tracks of alpha particles emitted by radon itself are first revealed in the etching processes, so we avoided taking into account tracks from the alpha particles of the radon progeny. The tracks were counted using the semi-automatic track-counting system consisting of a CCD camera and a high-resolution monitor. The signal from the monitor was sent to the computer where the picture was digitalized and analyzed using the appropriate program (Figure 4).

RESULTS AND COMMENTS

Table 2 shows the results of indoor radon concentration measurements performed in the spring of 2004 in several cities in Serbia.

Table 2. Results of indoor radon concentration measurements

Measurement location	Indoor radon concentration (BQM^{-3})
Arandelovac	33(6)
Zaječar	35(6)
Detector 25	29(5)
Bor	22(4)
Podgorica	21(4)
Pančevo	29(5)
Bor	26(5)
Pirot	35(6)
Pirot	33(6)
Pirot	31(6)
Niška Banja	30(5)
Vladičin Han	31(6)
Trstenik	27(5)
Detector 21	31(6)
Detector 12	27(5)
Niška banja	21(4)
Niš	30(5)
Beograd	27(5)

Radon concentrations were obtained on the basis of measured track density and use of the equations (5) and (7). It is obvious that the number of detectors, which were sent back, is smaller for the factor two compared to the number of the delivered detectors. So the first problem with these results is poor statistics. In future research, it is necessary to

have much more diverse measurement locations. Fortunately, the results shown in Table 2 are accepted as an intervention level for indoor radon concentration in Serbia and Montenegro. Because of the simplicity of the method described for radon measurements and relative low costs for assembling the laboratory for that purpose, it is possible to educate teachers and students in secondary schools to do independently all detection processes described in this paper. It is the way to get the first radon map of the territory of Serbia and continue monitoring of the variations in indoor radon concentrations as one of the most important procedures in the environmental protection.

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MERENJE RADONA ČVRSTIM DETEKTORIMA TRAGOVA: PRIMENA U SREDNJIM ŠKOLAMA

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U radu je opisan metod merjenja radona čvrstim detektorima tragova. Metoda je pogodna za simultano merenje na velikom broju lokacija što omogućava mapiranje radona. Urađen je probni eksperiment tako što je profesorima u više mesta u Srbiji podeljen određen broj detektora tipa CR-39. Detektori su eksponirani u periodu od tri meseca (proleće 2004. god.) na različitim lokacijama u nekoliko gradova u Srbiji. Nakon toga su poslani u Niskofonsku laboratoriju Instituta za fiziku, Zemun gde je izvršen hemijski tretman i analiza eksponiranih i razvijenih detektora. Preliminarni rezultati su prikazani u radu.