



Calibration of Solid State Nuclear Track Detectors CR-39 for radon study in a high concentration underground mines

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ABSTRACT

The Brazilian National Commission for Nuclear Energy (CNEN) establishes a reference level of 1000 Bq m⁻³ of radon concentration in underground mines. Solid State Nuclear Track Detectors – SSNTD is considered the main method of analysis in radon research. The methods and instrumentation used to determine radon concentration needs to be validated by calibration to ensure reliable results. This work aims to determine the calibration factor of CR-39 detectors exposed to well known radon concentrations produced in closed systems by sources of Ra-226 with activities of 3.379 kBq and 0.483 kBq, which are NIST sources of activities and emanation factors. The detectors were exposed to six different levels of exposure. The conversion factor between tracks density and exposure obtained was $K = 52.028 \pm 0.752$ [(tracks density.cm⁻²) / (kBq.d.m⁻³)]. As object of application of this work, mines were chosen with high radon concentrations reported by Santos (2015) in special a pegmatite mine located in the northwest of Minas Gerais. This mine presented radon concentration much higher than the recommended by the Brazilian national standard NN.CNEN.4.01, up to 7384 ± 517 Bq m⁻³.

Keywords: radon, CR-39, underground mines.

1. INTRODUCTION

The main source of radiation exposure in underground mines is ^{222}Rn ($t_{1/2}$ 3.83 d), a member of ^{238}U decay series, ^{219}Rn ($t_{1/2}$ 3.92 s), from ^{235}U decay series and ^{220}Rn ($t_{1/2}$ 54.5 s) that belongs to ^{232}Th decay series. The parent radionuclide of ^{222}Rn is ^{226}Ra ($t_{1/2}$ 1622 y), for ^{220}Rn is ^{224}Ra ($t_{1/2}$ 3.64 d) and for ^{219}Rn is ^{223}Ra ($t_{1/2}$ 11.1 d) [1]. ^{222}Rn is an alpha emitter with energy of 5.49 MeV and the most important radioisotope of radon in terms of radiation exposure and the subject of this paper. From now on, when it is written radon, refers to ^{222}Rn .

Alpha monitoring by Solid State Nuclear Track Detectors (SSNTD's) is considered the main method of measuring radon in the environment. SSNTD's can be used for measuring heavy particles as alpha particles and fission fragments. The system functions through air diffusion between the chamber and the environment, being, therefore, considered a passive method [2].

SSNTD have a number of advantages over other methods, such as low cost for acquisition and maintenance cost, absence of sensitivity to visible light and gamma radiation, good detecting efficiency, definitive record of track, efficiency stability over time, possibility to distinguish tracks due to the presence of protons, alpha particles and fission fragments. In addition, tracks recorded by the same type of particle, but with different energies, can also be distinguished [3].

Alpha-particle-sensitive materials often used are the polymers LR 115 (cellulose nitrate, $\text{C}_5\text{H}_8\text{O}_9\text{N}_2$), Makrofol, Lexan (polycarbonate, $\text{C}_{16}\text{H}_{14}\text{O}_3$) and Columbia Resin 39 (CR-39) (allylic diglycol carbonate, $\text{C}_{12}\text{H}_{10}\text{O}_7$) [4]. The CR-39 detector is a colorless and tough plastic with density of 1.30 g/cm^3 . It has become the most used tracks detector for monitoring radon in the environment, due to its high sensitivity and stability to ionization against various environmental conditions and its high degree of optical clarity [2].

CR-39 detectors are widely used for measurements of ^{222}Rn in houses, offices, underground mines, caves, water and exhalation circuits. The greatest advantage of CR-39 over other SSNTD is its efficiency. This detector is totally amorphous, providing regular tracks. CR-39 has a good contrast between the tracks and the detector, making it easier to observe this contrast in an optical microscope. In addition, CR-39 is a plastic very resistant against damage caused in environmental conditions, such as temperature, humidity, corrosive action of pollutants, among other [4].

The measurement of radon using this technique is based on simple methodology, which will be describe latter. However, it is difficult to maintain the measurements quality that should be assured by reproducibility and repeatability of the process. Several factors directly affect the results like detector material, environmental variables, tracks processing and counting, influence of the manufacturing process on detector sensitivity, storage conditions and time of use. In addition, changes in chemical etching and the efficiency of tracks counting must be considered [2]. Most effects of these factors can be overcome with the aid of the International Intercomparison exercise of radon/thorium monitors, ensuring the analyses quality. This intercomparison gathers results of different groups of different laboratories that perform the same activity and compare with each other [2].

A conversion factor obtained in calibrated systems allows converting tracks density to radon concentration, which is the number of tracks per unit area, after subtracting the background. The application of SSNTD in radon studies requires, as part of the quality assurance program, periodic revisions by re-calibration and participation in intercomparison programs [2].

CR-39 detectors should be calibrated and have the proportionality factor estimated, prior to its use and it is a very important tool for radon dosimetry environmental studies [5,6], since there is correlation between exposure of radon and its progeny and lung cancer, as shown in epidemiological studies [7,8,9]. Lung cancer is the second most common type of cancer in humans with more than 2 million cases in the world for year [10]. World Health Organization (WHO) recognizes radon as a carcinogenic factor [11] and, together with its progeny are responsible for more than 50% of the total radiation dose from natural sources. Thus, in recent years there has been a great scientific interest in the monitoring of this gas [7,8].

The Natural Radioactivity Laboratory (LRN) research group from the Nuclear Technology Development Center (CDTN) works with CR-39 detectors, specially towards the measurements of radon in underground mines. The mineral processing releases long and short lived radionuclides from uranium and thorium decay, which are considered the main source of exposure of miners to radiation. This exposure comprises internal exposition to short lived decay products of radon in the air and external exposition to gamma radiation from the walls [8].

Inside mines, radon present in mineral body is emanated through interstices present in grains or in fracture in the rocks. Radon is transported by molecular diffusion or convective flow reaching the galleries inside the mines.

Radon exhalation rate varies according to differences in pressure, degree of dilution, and porosity [8]. Radon concentrations in the air change substantially in the atmosphere of underground mines depending on the content of uranium, thorium and radium present in the geological formation, the type and daily activities of the mine, such as the working conditions and especially the degree of ventilation.

After exuded, radon migrates through the ventilation systems and forms its short half-life decay products: ^{218}Pb , ^{214}Pb , ^{214}Bi and ^{214}Po [11]. These radionuclides link to the aerosol particles present in the air, forming structures called attached radon progeny. Another part of the progeny, called unattached particles, stays in the air without attaching to particles. When inhaled, attached and unattached particles can be deposited in the respiratory system, especially in the upper respiratory tract, where they decay.

In this context, the objective of this work was to determine the calibration factor of CR-39, for LRN/CDTN and apply this factor to radon measurement in underground mines with high concentrations of radon. These measurements are part of the Radon Project headed by the Brazilian National Commission for Nuclear Energy (CNEN).

2. MATERIALS AND METHODS

A CR-39 detector, size 1x1.5 cm, 1 mm thickness, was attached to the diffusion chamber cover, a cylinder 2 cm high and 4 cm diameter with three air inlets. CR-39 remains inside the diffusion chamber along all the exposure time and are taken off for chemical etching process. To prevent radon decay products and dust inside the chamber, a filter paper with the same diameter was fixed on the chamber cover [13]. For transportation, CR-39, inside the diffusion chamber, are packed in aluminum foil for avoid cosmic radiation.

The alpha particles produce damage from their Coulomb interactions with the atoms in CR-39. These damages are of the order of 30 to 100 Å and are known as latent tracks [4]. One of the

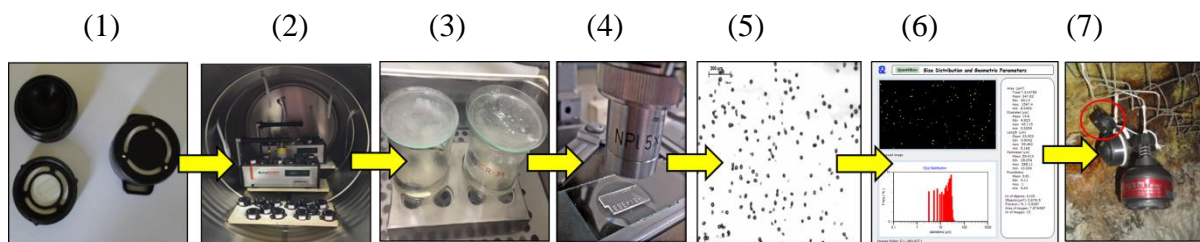
principles necessary to track is the Specific Energy Loss ($-dE/dx$, where E is the particle energy and dx is the middle depth).

The charged particle transfer its energy along the path as it enters the polymer. This loss of energy has a minimum required specific value, so the damage is severe and becomes traceable [4].

Some detectors are used for the background measurements at each exposure application. All detectors utilized in this work were kept together in a very low background room.

The entire process of calibration and measurement is shown in Figure 1. The process consists on (1) assembly of CR-39 detectors devices; (2) exposure of CR-39 in specific containers; (3) chemical etching process; (4) imaging acquisition; (5) counting of tracks; (6) processing for density acquisition and determination of the calibration factor and (7) application of the method and calibration factor in underground mines.

Figure 1: CR-39 calibration full process.



Source: Authors.

2.1. Exposure

For the calibration procedure, the radon detectors were submitted to six different levels of exposure, called Circuits I, II, III, IV, V and VI. The technical characteristics of each circuit varied according to the level of exposure required for the calibration. The technical information of each circuit is presented in Table 1.

Table 1: Technical information of calibration circuits.

Circuits	Source Activity of ^{226}Ra (kBq) [NIST standard]	Container Volume (L)	Total Experiment time (d)	Expected exposure (kBq.d.m ⁻³)
I	3.379	210.5	3.038	48.8
II	0.483	101.1	1.252	5.2
III	*	*	*	3.2
IV	*	*	*	14.8
V	3.379	210.5	2.00	32.1
VI	3.379	210.5	1.750	24.1

* Circuits III and IV were mounted in the National Institute of Radiation Science/Japan (NIRS) for international intercomparison programs where they have irradiated several other detectors for the LRN/CDTN laboratory. NIRS did not provide sources information and volume of the calibration chamber. The exposition time in each experiment in Japan was about 100 h.

Each circuit had twenty five CR-39-SSNTDs; two AlphaGUARD (Saphymo GmbH, model PQ2000PRO) detectors were also used as reference. Circuits I, V and VI used a container of 210.5 L, the two AlphaGUARD detectors and the 3.379 kBq ^{226}Ra source were positioned inside the container. In sequence, all operating conditions were verified, and source was opened. Then, the container was closed and kept airtight until the end of the experiment. Previously to the experiments, the source of ^{226}Ra (Pylon) had been maintained unchanged for more than thirty days until the secular equilibrium condition between ^{226}Ra and ^{222}Rn was established.

Circuit II present some peculiarities, ^{226}Ra (Saphymo) of activity equal to 0.483 kBq m⁻³ source was installed externally to the container and it was connected by means of Tygon hoses, which were installed in a glass bulb that needs to be kept moist all the time. Prior to experiment initiation, the background was determined, all the detectors (CR-39 and AlphaGUARD) were positioned and the circuit was closed and isolated from the source. Following the background measurement, the series source was placed in the circuit and the pump was turned on for 15 min to remove all ^{222}Rn sources. After that, the container valves were closed and the hoses disconnected. The container remained undisturbed until the end of the experiment.

Circuits III and IV were assembled by the intercomparison provider (NIRS/Japan) and, therefore, NIRS procedure was followed [15]. Intercomparison of results was carried out by NIRS of Japan, for a better certification of the calibration results. In this program, detection devices were prepared and sent to Japan for exposure at two known levels of radon concentration by the laboratory intercomparison provider. After experimental procedures, the detectors were taken back to LRN/CDTN for processing and analysis and the results obtained for ^{222}Rn concentration were compared between the two institutions.

2.2. Processing and determination of the calibration factor

After the exposures, the CR-39 detectors were removed from the detecting devices and etched in a solution of 6.25 M NaOH + 2% alcohol at a temperature of 75 °C and then washed for 20 min in running water [16]. The etching magnifies the tracks allowing them to be observed in a common optical microscope and to be counted, either manually or automatically for magnification of the tracks [5]. The background was determined on non-exposed detectors when subjected under identical conditions of etching [1].

Tracks count was done by means of an optical microscope (ORTHOLUX) with a 5x objective lens coupled to a DFC295 camera (Leica Microsystems Ltd.). For each detector, 15 images of the optical field were made with LAS V3.8 software, which were treated by the QUANTIKOV software [14] with a 0.003 μm filter and all standardized functions, providing the mean tracks density per cm^2 .

The concentration of ^{222}Rn (kBq m^{-3}) in the containers were measured by AlphaGUARD detectors. After the time sent in the procedure, exposure E value (kBq d m^{-3}) was obtained, where kBq is the activity concentration of radon, d is the length of exposure expressed in days and m^{-3} is the volume of studied place. The calibration factor k [(number of tracks. cm^{-2})/(kBq d m^{-3})] corresponds to the angular coefficient ($d = k.E$, where d is number of tracks per cm^2 of detectors present in the containers).

After determination of the calibration factor, CR-39 detectors were applied in underground mines measurements. Ten points were selected along the entire length of the mine, from air entry to exhalation area; in each point was installed one CR-39 detector device. CR-39 detectors were

exposed for approximately three months, according to the long-term measurement protocol of the US. Environmental Protection Agency [1]. After this time, the detectors were removed and developed for counting the tracks in order to obtain the radon concentrations concentrations using the process developed by LRN/CDTN [16].

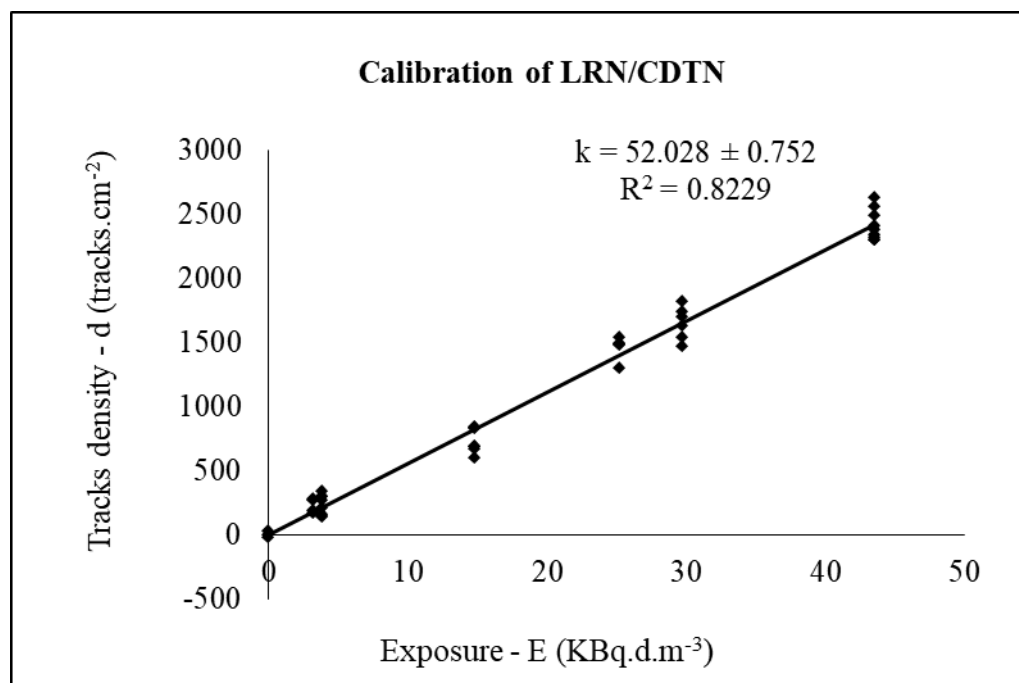
Santos (2010) reported high concentration of radon for the four mines selected to be studied in this project, which are located throughout Brazil and were named mines A, B, D and E. Mine A (Mine A1 and A2) is an agalmatolite deposit located at a ferriferous formation and present granite in its composition. Mine B is a coal mine, composed by arenito-aquifero, silito and coal gallery. Mine D is a tourmaline deposit in pegmatites located in a region with metamorphic rocks with quartzites, shales, amphibolites and filitos. Mine E is a scheelite deposit with main composition composed by silicate calcining, sheelita and calcium silicate [12].

The geological set of the mines monitored were described by a geologist. The local and visual data were registered at the local. Samples of rocks were collected for determining the activity concentration of the natural gamma emitters ^{226}Ra and ^{228}Ra (^{232}Th) by gamma spectrometry with HPGe detector and associated eletronic in the LRN/CDTN. Uranium concentrations were determined by Neutron Activation Analysis through Delayed Fission Neutrons. The neutron activation technique was carried out at CDTN using the TRIGA IPR-R1 research reactor. All the procedures followed the protocols of each laboratory responsible for the analyses.

3. RESULTS AND DISCUSSION

Proportionality factor between tracks density and exposure was determined by linear regression using MINITAB software. Uncertainty was obtained through the regression analysis performed with the 19 Degrees of Freedom corresponding to the 21 pairs of values, as shown in Figure 2.

Figure 2: Proportionality factor between tracks density and radon exposure determined in LRN/CDTN.



The intercomparison between LRN/CDTN and NIRS/Japan was performed under two different levels of exposure, high and low. Exposure of ²²²Rn made in low level presented average tracks of 224 cm⁻² and average radon's exposure of 4.0 ± 1.0 kBq d m⁻³ by LRN/CDTN and 3.2 kBq d m⁻³ by NIRS/Japan. Exposure made in high level presented average tracks of 737 cm⁻² and average radon's exposure of 13.2 ± 1.8 kBq d m⁻³ by LRN/CDTN and 14.8 kBq d m⁻³ by NIRS/Japan. Corresponding exposure was calculated using the proportionality factor determined by the LRN/CDTN and the NIRS/Japan reference exposure. The intercomparison provider, NIRS, did not provide the uncertainties, which is negligible compared to the participating laboratories. All the reported exposures are reference values traceable to the National Institute of Standards Technology of the United States (NIST).

The average radon concentration from mines with high concentrations, determined using CR-39 detectors are shown in Table 2. These mines present high values of radon activity concentrations comparing to the CNEN limits, which is 1000 Bq m⁻³ for the working sites [17]. Mine D presented values above the national limit and mines A, B and E had at least one point above

the limit, thus some actions are recommended to decrease radon concentrations, improving the workers' health conditions, such as better ventilation and establishment of a radon monitoring program.

Table 2: Radon concentration in underground mines.

Mine	Operation Status	Average radon concentration in air (long term measurements) (Bq m ⁻³)
A-1	Closed	676 ± 64
A-2	Open	778 ± 70
B	Open	985 ± 98
D	Closed	4153 ± 343
E	Open	779 ± 73

The activity concentrations (Bq/kg) of the radionuclides ²²⁶Ra (²³⁸U), ²²⁸Ra (²³²Th) and ²³⁸U, present in the rock samples collected in the studied mines are shown in Table 3.

Table 3: Minimum and maximum activity concentrations of the radionuclides ²²⁶Ra (²³⁸U) and ²²⁸Ra (²³²Th) in the studied underground mines.

Mine	Activity concentrations (Bq/kg) of the radionuclides*				²³⁸ U (Bq/kg)	
	²²⁶ Ra (²³⁸ U)		²²⁸ Ra (²³² Th)		Min	Max
	Min	Max	Min	Max		
A	40.9 ± 0.5	120.1 ± 0.6	66.8 ± 0.3	187.0 ± 0.7	98.4 ± 24.6	209.0 ± 36.9
B	195.5 ± 0.8	517.4 ± 1.7	5.8 ± 0.3	50.7 ± 0.4	196.7 ± 24.6	1057.4 ± 49.2
D	10.8 ± 0.3	297.4 ± 1.0	0.4 ± 0.1	28.0 ± 0.3	12.3 ± 12.3	909.9 ± 49.2
E	8.3 ± 0.2	59.8 ± 0.4	3.2 ± 0.1	8.3 ± 0.1	12.3 ± 12.3	110.7 ± 24.6

*In each studied mines was collected more than one sample and the values refers to it.

The values to worldwide average concentration of ²²⁶Ra and ²³²Th, just for comparison, reported by UNSCEAR (2000) are 32 and 45 Bq/kg, respectively [18]. The results were above to

the recommended limit in all studied mines for ^{226}Ra and in mine A and B for ^{232}Th . Therefore, these mines should be studied with more details.

4. CONCLUSION

Calibration factor was determined utilizing the Calibration Systems developed by LRN/CDTN, obtaining a value of $k = 52.03 \pm 0.75$ [(tracks.cm⁻²)/(kBq d m⁻³)]. As a result, this factor can be used to determine radon concentration activity in air, in places like homes, workplaces, underground mines and other. The same factor can be used in exhalation circuits and in research projects carried out by the LRN/CDTN with guaranteed quality, as long as the same processing conditions and restricted control are used.

The results of the inter-comparison program with NIRS/Japan showed very good agreement between the two points of the calibration curve obtained by irradiation and the respective values provided by NIRS, which allow concluding that the experiments led to a very reliable Calibration Factor that can confidently be used to monitor underground mines and in other related projects.

The monitoring of ^{222}Rn in underground mines in compliance with CNEN standard is important for safety and security of the workers in underground environments. The control and monitoring of the concentration of these mines is necessary, since levels above the reference limit were found in some mines. The measurement of mine D was ten times higher than the reference level established by CNEN, then, this mine requires special attention. The other mines need to be monitored for safety and security of the workers, as the mines conditions change all the time.

This study also demonstrated that SSNTD's are an adequate tool to measure radon concentration in air especially because there was not saturation of the detectors, being efficient for application in underground mines.

This work suggests other measurements in the mines for comparison purposes. Calibration of detector CR-39 in other higher and lower values of exposure are also suggested to improve the calibration factor.

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