*2013 International Nuclear Atlantic Conference - INAC 2013*
*Recife, PE, Brazil, November 24-29, 2013*
*ASSOCIAÇÃO BRASILEIRA DE ENERGIA NUCLEAR - ABEN*
**ISBN: 978-85-99141-05-2**

# EFFICIENCY CALIBRATION FACTOR FOR RADON EXHALATION IN ROCKS BY USING CR-39 SSNTD AND THE "SEALED-CAN" TECHNIQUE

**Ademar O. Ferreira*[1], Brigitte R. S. Pecequilo[2] and Reginaldo R. Aquino[3]**

Instituto de Pesquisas Energéticas e Nucleares (IPEN / CNEN - SP)
Av. Professor Lineu Prestes 2242
05508-000 São Paulo, SP
aoferreira@ipen.br[*1] brigitte@ipen.br[2], raquino@ipen.br[3]

## ABSTRACT

In this work, the calibration factor for CR-39 solid state nuclear track detectors was determined, for furher determination of radon exhalation rate of undamaged geological rocks samples using the sealed can-technique. Four CR-39 detectors within and without diffusion chambers were exposed to known radon activities concentrations and different integration periods inside a stainless steel calibration chamber with an inner volume of 0.0144 cubic meters and a calibrated source of radium-226 operating as an intermittent source. The activities varied from 0.67 kBq.m$^{-3}$ to 165 kBq.m$^{-3}$ and the exposure times from 20 h to 316.5 h. The mean calibration factor and its standard deviation assumed for further use was 2.5 ± 0.1. The results show a good reproducibility of measures for a broad spectrum of radon activities concentrations.

## 1. INTRODUCTION

Almost 46% of exposure to mankind by natural sources is due to inhalation of radon and its short-lived decay products. Radon is an inert gas with three natural isotopes occurrence, $^{222}$Rn, $^{219}$Rn and $^{220}$Rn, originated from the natural radioactive series of $^{238}$U, $^{235}$U and $^{232}$Th, respectively [1]. In terms of radiological concern, considering the short half-lives of $^{220}$Rn (55.6 s) and $^{219}$Rn (3.96 s) and that $^{235}$U represent only 0.71% of the natural uranium, the major contributor to exposure is the inert gas $^{222}$Rn (3.8 d) (in this paper, radon), unless for places with high $^{232}$Th concentrations when $^{220}$Rn contribution will be significant [2].

The release of radon from geological samples is considered to be both by emanation or exhalation. Radon emanation is defined by Sakoda [3] as escape of a radon atom from a Ra-bearing grain into pore space [1]. The radon in pore can leave it by diffusion or together with a flow of air or water, this processes of release radon from material to air is named exhalation [4, 1].

Exhalation from building materials or recovering materials is a potential source of radon in the indoor environment, so, the knowledge of its contribution to the dose exposure is very important. However, it is not possible to determine the radon exhalation rate simply from the activity concentration of $^{226}$Ra. Rather then, one must measure radon exhalation rates directly from the surface of the material [5].

Radon exhalation measurements are usually based on the principle of hermetically enclosing the sample in a container and measuring the radon activity growth as a function of time inside

the container. In environmental studies, the most widely used technique for radon detection is the passive one with CR-39 solid state nuclear track detectors (SSNTD).

Several brazilian rocks were assessed for radon exhalation determination by using the "Sealed-Can Technique" [5, 6], a simple cylindrical polyethylene bucket with lid, tightly sealed, where the sample is placed on bottom and the CR-39 detector on top. After a predetermined exposure time, the bucket is open and the detector read. The correct radon activity concentration depends on a well-calibrated detector. The aim of this study is to determine the calibration factor for the CR-39 detector.

## 2. MATERIALS AND METHODS

### 2.1 Experimental procedure

In any physical measurement, the calibration factor (sometimes called efficiency factor) must be determined for the same geometry conditions as the studied samples. As the samples of interest are commercial, unprocessed ones, with dimensions of 15 *x* 15 *x* 2 cm, in order to fit the samples for the "Sealed-Can Technique", a cylindrical polyethylene bucket with a diameter of 25 cm and height of 26.5 cm was used.

The calibration factor was determined in a stainless steel can, similar to the cylindrical polyethylene bucket, with an inner (useful) volume of 0.0144 $m^3$, adapted to (1) receive and hold known amounts of radon, (2) allow that CR-39 detectors be placed inside. On the upper lid of the calibration chamber (Fig.1) three outlets, pneumatic push-to-connect fittings, (two for gas inlet and outlet and one for the leak valve) were adapted. Both outlet and inlet gas fittings were needle valves.

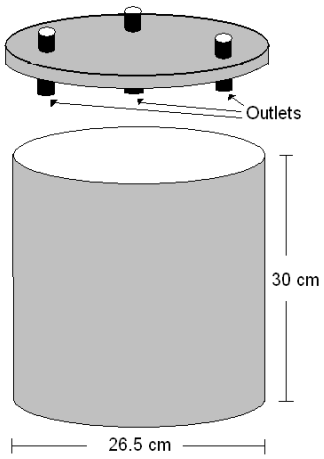


**Figure 1: Calibration chamber for calibration factor for CR-39 indoor radon detector.**

In order to evaluate the uniformity of radon in the chamber and the reproducibility of measures, four detectors were placed inside the chamber, at different locations (Fig. 2). Two CR-39 detectors inside NRPB/SSI diffusion chambers [7] were positioned at the bottom (position 1) and top (upper lid) of the calibration chamber (position 2). On top, at position 3, a bare CR-39 detector and at position 4, a CR-39 from a different manufacturer, inside a

NRPB/SSI diffusion chamber. Detectors at positions 2, 3 and 4 on the upper lid were equidistantly placed.

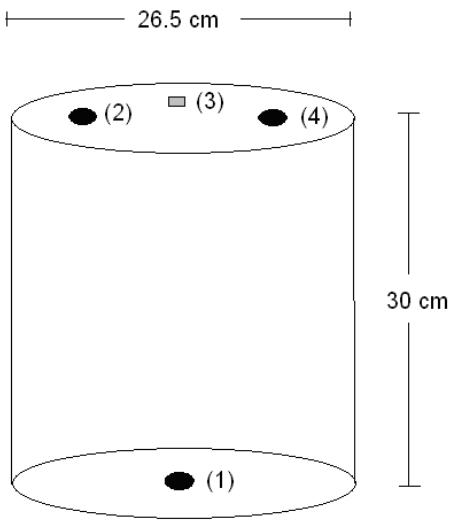


**Figure 2: Diagram of detectors inside the calibration chamber.**

The radon calibrated source was a flow-through type (Pylon model RN-1025), with nominal activity of 107.318 kBq of $^{226}Ra$, operating as an intermittent source (the quantity of radon gas accumulated within the source cavity is transferred to the calibration chamber).

Known quantities of radon activities concentrations were transferred to the calibration chamber that was sealed for a predetermined integration time, as in Table 1.

**Table 1:** Radon activity concentration transferred to chamber for calibration factor determination and their respective times of integration.

| Activity ($kBq.m^{-3}$) | Radon integration time (h) |
|---|---|
| 0.67 ± 0.03 | 316.5 |
| 3.5 ± 0.1 | 72 |
| 10.5 ± 0.4 | 96 |
| 20.9 ± 0.8 | 96 |
| 31.3 ± 1.3 | 96 |
| 41.7 ± 1.7 | 96 |
| 52.2 ± 2.1 | 96 |
| 72.6 ± 2.9 | 44 |
| 83.1 ± 3.3 | 24 |
| 104.0 ± 4.2 | 22.63 |
| 124.2 ± 5.0 | 24 |
| 145.1 ± 5.8 | 20 |
| 165.0 ± 6.6 | 24 |

For every radon activity transferred to the calibration chamber, a new set of CR-39 detectors was used.

After exposure, the detectors were etched for 5.5 hours with a KOH 30% solution at 80°C [8] and read under a Zeiss optical microscope [9].

### 2.2 Determination of the calibration factor

A calibration factor was obtained for each activity value transferred from the source cavity to the calibration chamber, for the specific time of integration using eq. 1, modified by Orlando [9]

$$F = \frac{D}{C_{Rn}.t} \tag{1}$$

with:

F = detector calibration factor (tracks.$m^3$).($cm^{-2}$.$Bq^{-1}$.$h^{-1}$);
D is= tracks density (tracks.$cm^{-2}$),
$C_{Rn}$ = radon concentration (Bq $m^{-3}$)
t = exposition time (hours)

The uncertainty of the detector calibration factor ($\sigma_F$) was calculated from equation 2, considering the uncertainty of track density ($\sigma_D$) and the uncertainty of radon concentration ($\sigma_{C_{Rn}}$).

$$\sigma_F = F\sqrt{\left(\left(\frac{\sigma_D}{D}\right)^2 + \left(\frac{\sigma_{C_{Rn}}}{C_{Rn}}\right)^2\right)} \tag{2}$$

The calibration factors F1, F2, F3 and F4, for the CR-39 detectors 1, 2 3 and 4 were calculated as the arithmetic mean of the individuals factors obtained for each radon activity concentration to which they were exposed, with a standard deviation of ±1σ confidence level.

## 3. RESULTS

The tracks densities for each one of the four CR-39 detectors, placed in the bottom (1) and on the lid (2, 3 and 4) of the calibration chamber (Fig. 2), normalized by time and related to the radon activities concentrations are shown in Fig. 1.

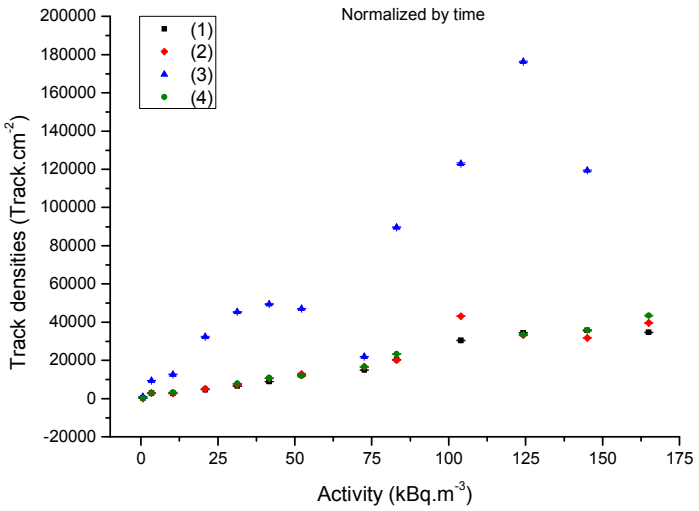


**Figure 3: Normalized track densities as a function of radon activities concentrations.**

The good correlation between tracks densities and the respective activities indicates an acceptable reproducibility of the results, considering the wide range of activities (from 0,67 kBq.m$^{-3}$ to 165 kBq.m$^{-3}$).

In Fig. 4, 5 and 6 the tracks densities of the top placed detectors detectors 2, 3 and 4 are related to the bottom detector 1, respectively.

Detectors 1, 2 and 4 were exposed placed inside diffusion chambers, whereas detector 3 was exposed plain.

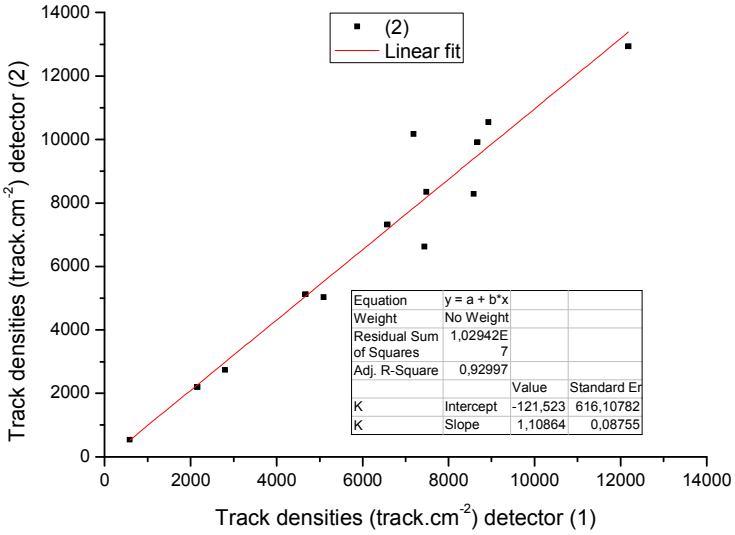


**Figure 4: Track densities for top detector 2 as a function of track densities for bottom detector 1.**

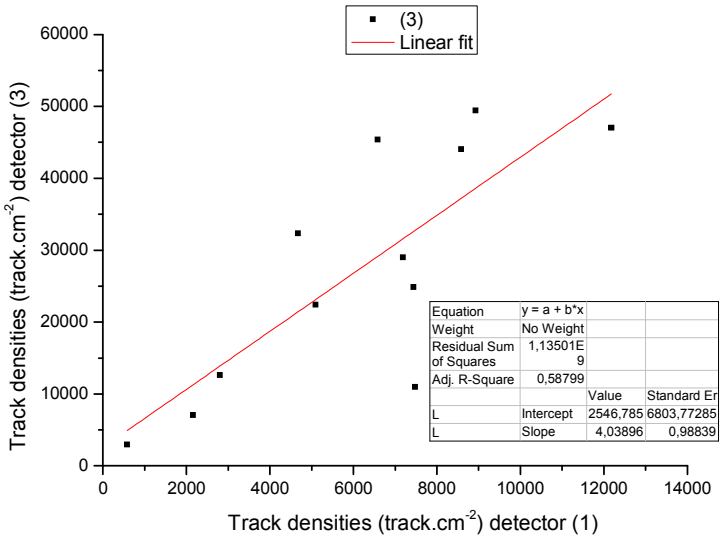


**Figure 5: Track densities for top detector 3 as a function of track densities for bottom detector 1.**

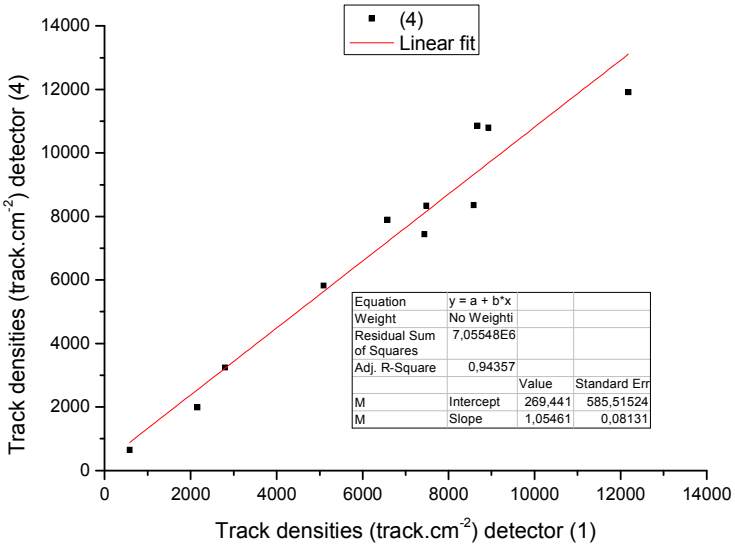


**Figure 6: Track densities for top detector 4 as a function of track densities for bottom detector 1.**

Fig. 4 and 6 show the best fit, meaning that, even the detectors being from different manufacturers, their efficiencies were similar. Also, the good correlation between the bottom

and top detectors indicates that radon was uniformly distributed in the entire chamber. The worst correlation (Fig 5) can be induced by the fact that detector 3 was exposed with no diffusion chamber. However, the results can be used, as they present statistically acceptable values.

The calibration factors F1, F2, F3 and F4, calculated from eq. 1 for CR-39 detectors 1, 2, 3 and 4, respectively, are presented in Fig. 7.

In Fig 7, the F3 values are higher than the others ones, also varying within a larger range, from 3.44 ± 0.01 (tracks.m$^3$).(cm$^{-2}$.Bq$^{-1}$.h$^{-1}$) to 16.22 ± 0.03 (tracks.m$^3$).(cm$^{-2}$.Bq$^{-1}$.h$^{-1}$). Considering the value 28.08 ± 0.05 (tracks.m$^3$).(cm$^{-2}$.Bq$^{-1}$.h$^{-1}$) as an outlier, the mean value with its standard deviation is 11.80 ± 0.02 (tracks.m$^3$).(cm$^{-2}$.Bq$^{-1}$.h$^{-1}$).

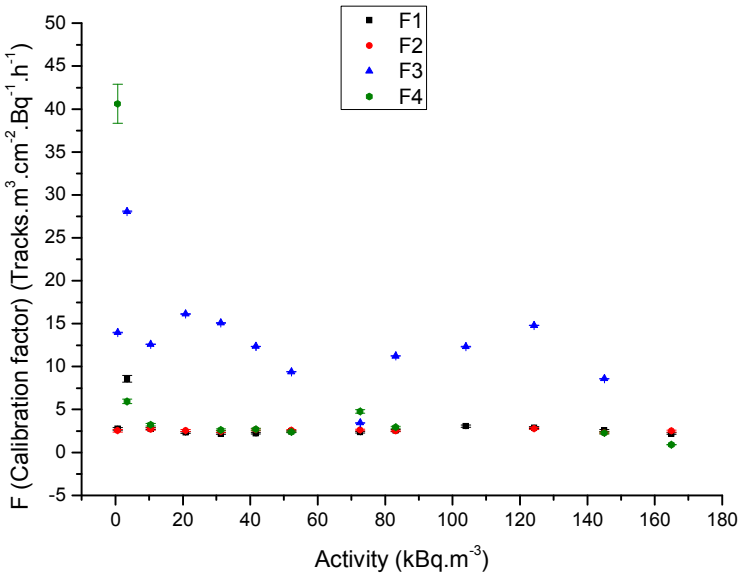


**Figure 7: Calibration factor for all detectors exposed in the calibration chamber**.

As in the assessment of radon exhalation rates, the detectors must be exposed in diffusion chambers, the calibration factor is presented in Fig. 8, considering only the three detectors, 1, 2 and 4.

Assuming the values of 8.6 (tracks.m$^3$).(cm$^{-2}$.Bq$^{-1}$.h$^{-1}$) for detector 1 and 41 (tracks.m$^3$).(cm$^{-2}$.Bq$^{-1}$.h$^{-1}$) for detector 4 as outliers, the results are in the range from 0.9 (tracks.m$^3$).(cm$^{-2}$.Bq$^{-1}$.h$^{-1}$) to 5.9 (tracks.m$^3$).(cm$^{-2}$.Bq$^{-1}$.h$^{-1}$), with a mean value and standard deviation of 2.7 ± 0.8 (tracks.m$^3$).(cm$^{-2}$.Bq$^{-1}$.h$^{-1}$).

Finally, considering only detectors 1 and 2 from the same manufacturer, the results for the calibration factors are presented in Fig.9.

The two detectors of same origin show the best reproducibility with a mean value and standard deviation of 2.5 ± 0.1 (tracks.m$^3$).(cm$^{-2}$.Bq$^{-1}$.h$^{-1}$), even placed in different positions inside the calibration chamber. This value has the same order of magnitude as Orlando [9] suggested value of 2.8 ± 0.2 (tracks.m$^3$).(cm$^{-2}$.Bq$^{-1}$.h$^{-1}$) for the same detector in environmental measures (out of a sealed can), and Matiullah [10] value of 2.56 (tracks.m$^3$).(cm$^{-2}$.Bq$^{-1}$.h$^{-1}$) for the same geometry.

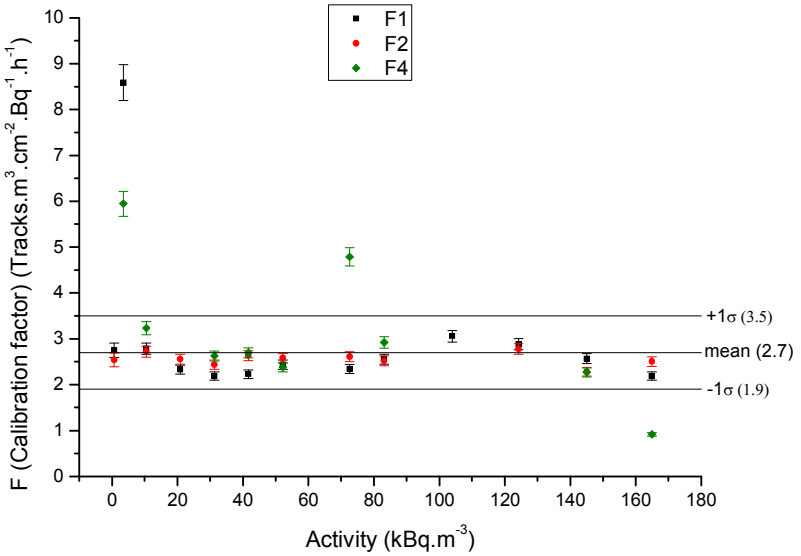


**Figure 8: Calibration factors for detectors of different manufacturers exposed within diffusion chambers.**

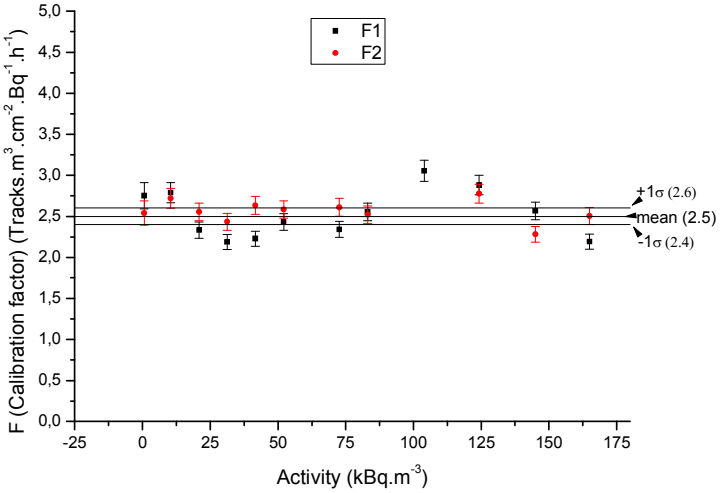


**Figure 9: Calibration factors for detectors from the same manufacturers exposed within diffusion chambers**

## 3. CONCLUSIONS

In this work, calibration factors for CR-39 detectors were determined using a calibration chamber and the "Sealed Can Technique", for further determinations of radon exhalation rates in rocks.

The good correlation between the detectors tracks densities and the radon activities concentrations indicates the reproducibility of measurements. The worst result ($R_2$ = 0,59) was obtained for the bare exposed CR-39 detector, reinforcing the necessity of the use of a diffusion chamber for an uniform distribution of radon progeny.

The calibration factors measured for detectors inside the diffusion chamber show an acceptable statistical distribution. The largest deviation was obtained for detector 4, from a different manufacturer that detectors 1 and 2, meaning that for detectors of different origins it is necessary to determine their specific calibration factors.

As the purpose of this work is the direct application of the calibration factor in the determination of radon exhalation rocks using CR-39 detectors type 1 and 2, the calibration factor assumed was 2.5 ± 0.1 $(tracks.m^3).(cm^{-2}.Bq^{-1}.h^{-1})$, in good agreement with literature values.

## ACKNOWLEDGMENTS

This work was conducted at the Environmental Radiometric Laboratory of IPEN and supported by CNEN and CNPq.

## REFERENCES

1. UNSCEAR – 2000, United Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), The 2000 Report to the General Assembly with scientific Annexes. New York: United Nations. (2000).
2. UNSCEAR – 1993, United Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), The 1993 Report to the General Assembly with scientific Annexes. New York: United Nations. (1993).
3. Sakoda A., Ishimori Y., Yamaoka K., A comprehensive review of radon emanation measurements for mineral, rock, soil, mill tailing and fly ash. Applied Radiation Isotopes 69. 1422-1435. (2011).
4. Faheem M., Matiullah,. Radon exhalation and its dependence on moisture content from samples of soil and building materials. Radiation Measurement 43. pp.1458-1462. (2008).

5. Ferreira, A, O., Avaliação da radioatividade natural em algumas rochas graníticas do estado do Paraná e sua utilização na construção civil, Tese de Doutorado apresentada ao Instituto de Pesquisas Energéticas e Nucleares. (2013).
6. Ferreira, A.O.; Pecequilo, B.R.S; Aquino, R.R. Application of a Sealed Can Technique and CR-39 detectors for measuring radon emanation from undamaged granitic ornamental building materials. Radioprotection (Paris. 1966), v. 46, p. S49-S54, (2011).
7. Pereira, Osvaldo L.S.P; Estudo comparativo das respostas de monitores de Rn-222 que utilizam o CR-39 como detector de traços de partículas alfa. Dissertação de Mestrado. UNICAMP (2010).
8. ZEISS, C., KS 100 Imaging System Release 3.0, (1997).
9. Orlando, C. Orlando, P., Patrizii, L., Tommasino, L. , Tonnarini, S., Trevisi, R. And Viola, P., A passive radon dosimeter suitable for workplaces. Radiat. Prot. Dosim., v. 102 p. 163–168. (2002).
10. Mautillah, Determination of the calibration factor for CR-39 based indoor radon detector. J. Radioanal Nucl Chem. DOI 10.1007/s10967-013-2451-9 (2013).