〈 Technical Note 〉

CONSTRUCTION OF AN ENVIRONMENTAL RADON MONITORING SYSTEM USING CR-39 NUCLEAR TRACK DETECTORS

GIL HOON AHN* and JAI-KI LEE[1]
National Nuclear Management & Control Agency
305-600, POX 114 Yuseong, Daejeon, Korea
[1]Dept. of Nuclear Engineering Hanyang University
17 Haengdang-Dong, Sungdong-Gu, Seoul, 133-791 Korea
*Corresponding author. E-mail : ghahn-nnca@kins.re.kr

Received December 7, 2004
Accepted for Publication May 31, 2005

An environmental radon monitoring system, comprising a radon-cup, an etching system, and a track counting system, was constructed. The radon cup is a cylindrical chamber with a radius of 2.2 cm and a height of 3.2 cm in combination with a CR-39 detector. Carbon is impregnated in the bodies of the detector chamber to avoid problem of an electrostatic charge. The optimized etching condition for the CR-39 exposed to a radon environment turned out to be a 6 N NaOH solution at 70ºC over a 7hour period. The bulk etch rate under the optimized condition was 1.14±0.03 μm h^{-1}. The diameter of the tracks caused by radon and its progeny were found to be in the range of 10~25 μm under the optimized condition. The track images were observed with a track counting system, which consisted of an optical microscope, a color charged couple device (CCD) camera, and an image processor. The calibration factor of this system is obtained to be 0.105 ± 0.006 tracks cm^{-2} per Bq m^{-3} d.

KEYWORDS : Radon, Radon Cup, Etching, CR-39, Solid State Nuclear Track Detector(SSNTD)

1. INTRODUCTION

Radon is the largest and most variable contributor of public exposure to radiation. It is estimated that the annual effective dose by radon and its progeny from the inhalation of air is about 50% of natural public exposure dose rate and prolonged exposure to high levels of radon can cause lung cancer [1]. In recent years, interest in this subject has been increasing rapidly in Korea because of news that the radon concentration of underground water in some regions and air of some Seoul subway stations is higher than action guideline level of other countries [2-4]. Measurement of radon exposure has gained added significance because of the increased potential for lung cancer caused by the combined effects of radon, air pollution, and smoking [5].

The environmental radon concentration is a function of time and climate conditions. To monitor radon, both active and passive techniques have been developed. Active methods are usually used for short-term measurements of radon and for detailed investigations of individual sites under inspection. Passive methods are more suitable for the assessment of radon exposure over long time scales and can be used for large-scale surveys at moderate cost. For that reason, many countries have performed large-scale radon surveys using passive monitoring devices, have assessed the public exposure dose rate from radon, and have adopted appropriate actions for protection against radon [6-9]. Therefore, the construction of reliable and inexpensive radon monitoring system to assess the radon exposure should be done first in Korea.

In this work, radon cup using solid-state nuclear track detector (SSNTD), which have the ability to integrate over multiple day-long intervals of time at dwelling and building is developed along with a track counting system. Additionally, the optimum etching condition is also found. The proposed system can be used for large-scale surveys of environmental radon.

2. EXPERIMENTAL METHODS AND RESULTS

2.1 Radon Cup

A radon cup used for radon exposure assessments is made up of detection material and detector chamber. A radon cup used over long time scales with large-scale surveys must be small, low cost, and easy to both handle

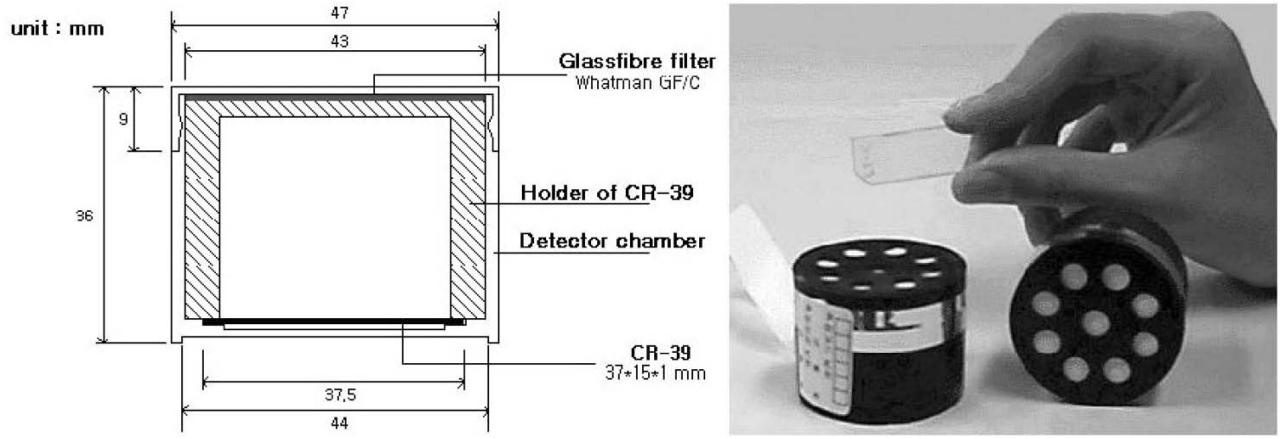

Fig. 1. The Radon Cup Developed in this Study

and read [10]. The sensitivity of a radon cup is dependent on the material and volume of the detector chamber, the position of the detection material in the detector chamber, and the filter that shuts out radon progeny.

2.1.1 Detection Material

The most popular member of the SSNTDs family, CR-39, was selected because of its good sensitivity, stability against various environmental factors, and high degree of optical clarity. Large sheets of CR-39 were supplied by Fukuvi Co., Ltd., Japan. Sheets of 0.9 mm thickness were used for robustness and to avoid the possibility of tracks on the back surface being detected by the image analyzer. These sheets were cut into rectangular shapes sized 1.5 cm × 3.7 cm, and one corner of the rectangle was removed to allow for proper orientation. A serial number was engraved on each element in Arabic numerals for ease of identification.

2.1.2 Detector Chamber

The detector chamber is a cylindrical cup of 2.2 cm in radius and 3.2 cm in height. Carbon is impregnated in the wall material, polypropylene, to enhance electrical conductivity and to avoid the problem of electrostatic charge. This cup is sealed with glass-fibre filter (Whatman GF/C, England) that discriminates short-lived thoron by delaying the entry of gases into the chamber, limits access of moisture, and blocks the entry of radon progeny and dust present in the ambient air [11-13]. The CR-39 on the bottom of the detector chamber is fixed by holder to reduce any error that might be caused by its movement. Radon enters the holder with a half-time for entry about 1 minute, which is short compared with the radon half-life of 3.82 days [11]. This means that the radon concentration inside the detector chamber quickly approaches that outside. It can be shown that the long-term average radon concentration inside the detector chamber is the same as that outside, despite any variations in the outside concentration. But the radon concentration may be overestimated because the short half-time for entry will allow some thoron to enter the detector [11]. The radon cup developed in this study is shown in Figure 1.

2.2 Track Etching System

The optimal use of any track detector is largely dependent on standardization of various etching parameters, such as the bulk etch rate (V_b) and track etch rate (V_t), both of which must be experimentally determined under suitable conditions. A set of systematic experiments was carried out to find the optimal etching condition. CR-39 samples were irradiated using two alpha source (2.4 MeV and 4.3 MeV) with 2π geometry. Irradiated CR-39 samples were etched in a NaOH solution, which is the most popular etchant and has been extensively studied [14]; varying concentrations of NaOH solution were used, from 3~10 N, at temperatures ranging from 50 to 80 ºC, during periods of 4 to 10 hours. After etching, the CR-39 samples were cleaned in running water for 20 minutes and dried flat between tissue wipes to remove the etchant and etch products from the surface of the detector. The optimum etching condition for the CR-39 used in this study turned out to be etching the CR-39 in a solution of 6 N NaOH solution at 70 ºC over a 7 hour period. Figure 2 shows the changes of the bulk etch rate with normality of an aqueous solution of NaOH at 70 ºC, and Figure 3 shows the one with temperature of an aqueous solution of NaOH at 6 N. It can be seen in these figures that the normality has a stronger effect than temperature, and reproducibility of the etching temperature and normality is important, because the bulk etch rates change rapidly with temperature above 60 ºC. Values of the ratio between the track radius and the thickness of the removed surface are plotted as a function of the normality and temperature of the NaOH solution in Figures 4 and

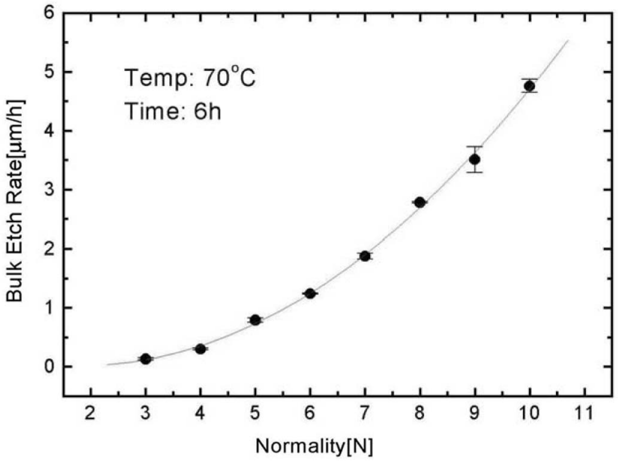

Fig. 2. The Change of Bulk Etch Rate with Normality of an Aqueous Solution of NaOH

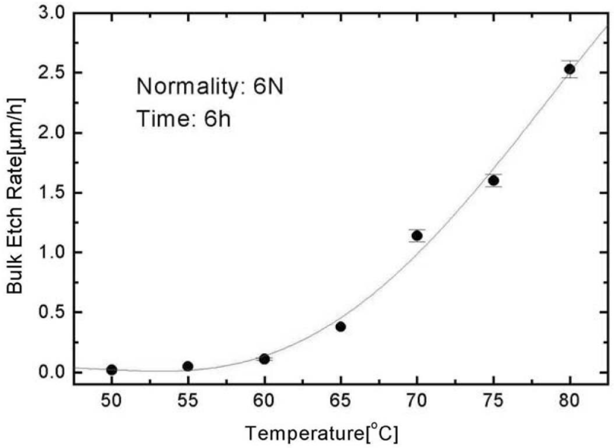

Fig. 3. The Change of Bulk Etch Rate with Temperature of an Aqueous Solution of NaOH

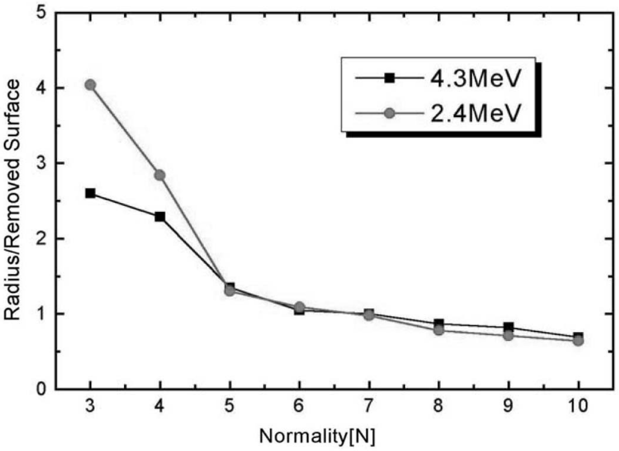

Fig. 4. Variation of the Ratio Between Track Radius and Thickness of Removed Surface as a Function of Normality of an Aqueous Solution of NaOH

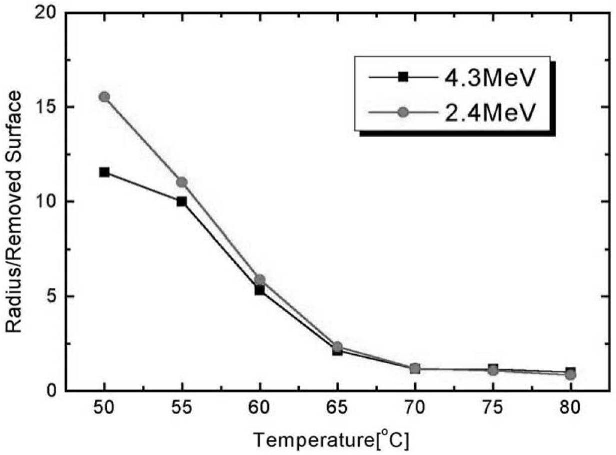

Fig. 5. Variation of the Ratio Between Track Radius and Thickness of Removed Surface as a Function of Temperature of an Aqueous Solution of NaOH

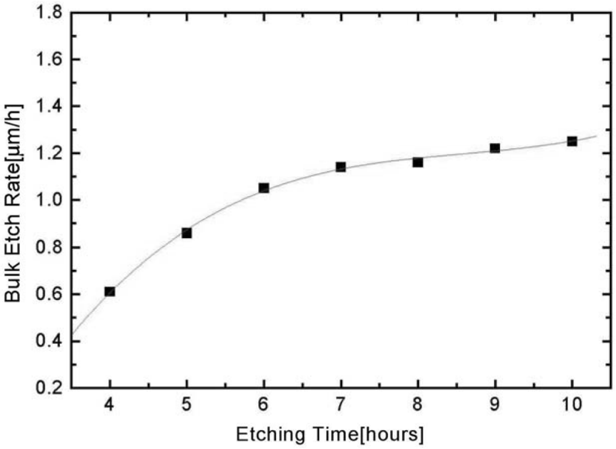

Fig. 6. Dependence of the Bulk Etch rate of CR-39 on the Etching Time

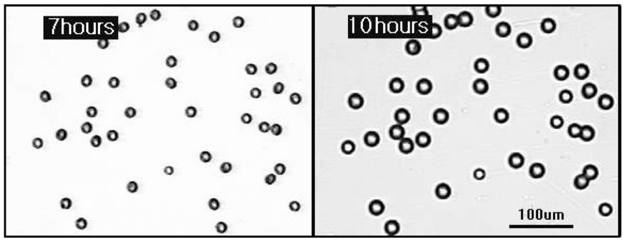

Fig. 7. Tracks of 4.3 MeV α-particle Produced in CR-39 for Different Etching Times

5. It is clear from these figures that the etching sensitivity (V_t / V_b) is high because the ratio between the track radius and the thickness of the removed surface is low in a solution of 6 N NaOH at 70ºC. The dependence of the bulk etch rate

on the etching time is shown in Figure 6. The bulk etch rate was found to be saturated after 7 hours. The bulk etch rate under the optimized condition was 1.14 ± 0.03 μm h^{-1}, and the diameters of the etched tracks were in the range of 10~25 μm. Figure 7 shows the appearance of the tracks of α-particle of 4.3 MeV produced in CR-39 for different etching times at a magnification of 149.

2.3 Track Counting System

The etched tracks were observed using an optical microscope fitted with an objective lens of 149 times magnification. At this magnification one counting field covers an area of 0.99 mm^2. The microscope image was viewed with a high-quality monochrome charge coupled device (CCD) TV camera, which is connected to a PC-based image analyzer (Image-Pro Plus version 4.0). The image analyzer displays images on a monitor. Tracks are counted automatically in 10 different fields around center of the detector (covering 9.9 mm^2). For unexposed detectors used for the assessment of background track density, 20 different fields were scanned. The tracks appeared as dark spots on a clear white background, and a grey-level threshold detection was performed to separate the tracks from the clear CR-39. Tracks were not accepted as genuine unless their areas and roundnesses ($\text{perimeter}^2/(4*\pi*\text{area})$) fell within the acceptance criteria. The upper and lower limits for area of acceptable tracks were 50 and 450 μm^2 and for roundness 1 and 1.8, respectively, where a roundness of 1 refers to a disc. The track counting system developed in this study is shown in Figure 8.

2.4 Calibration and Intercomparison

Calibration experiments were carried out to evaluate the relationship between the track density recorded and the radon concentration. Reliable measurements of radon concentrations with an integrating device depend sensitively on the soundness of the calibration procedure. Exposure to radon was done at the National Radiological Protection Board (NRPB, UK) radon chamber [15]. This facility has been the European regional reference laboratory for radon measurements under an intercalibration and intercomparison scheme organized by the IAEA. The radon level varied between 65kBq h m^{-3} and 397kBq h m^{-3}, similar to the exposure received in 2~11 months in a dwelling at the Korea average radon concentration of 50 Bq m^{-3}[16], with 12 radon-cups at 6 different level points. The corresponding track density for the CR-39 varied from 291.8±58 tracks cm^{-2} to 1763.7±88 tracks cm^{-2}. Figure 9 shows the relationship between track densities and radon concentrations. The straight line represents the least squares fit of the data. The calibration factor for each of the data points are presented in Table 1. A mean value obtained from the experiment was 0.105±0.006 tracks cm^{-2} per Bq m^{-3} d.

Before using this system for a large survey program, it is essential to test the limit of the application at higher exposure levels. For that reason, exposure to radon was done at the Korea Research Institute of Standards and Science radon chamber [17]. The radon level varied between 396kBq h m^{-3} and 795kBq h m^{-3} with 5 radon-cups at 4 different level points. Figure 10 shows the result of the linearity test. It is clear from this figure that the detector response is maintained to about 800kBq h m^{-3} with good linearity.

To ensure the quality of a radon measurement, it is important to compare different detectors exposed side by side. Although passive radon measurement techniques are simple in principle, it has been found that it is difficult in practice to maintain good quality control. AlphaGUARD, which is an active device and the radon-cup developed in this study were exposed together in a radon chamber at Hanyang University. The AlphaGUARD device is the centerpiece of a portable measuring system for the continuous determination of radon concentration, and uses the proven principle of the pulse ionization chamber [18]. The

Fig. 8. Track Counting System

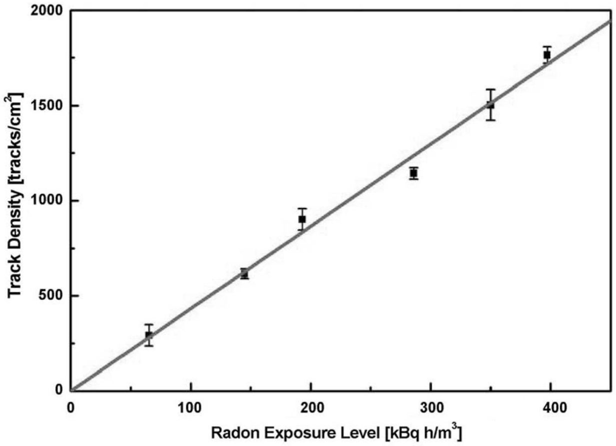

Fig. 9. Calibration of the Detector Against Radon Concentration. Damage to LWR Secondary-side Piping [1]

Table 1. Calibration Factors for Detectors Exposed in the Radon Cup Versus Radon Level

Radon level [kBq h m^{-3}]	Track density [tracks cm^{-2}]	Calibration factor[tracks cm^{-2} per Bq m^{-3} d]
65	291.8 ± 57	0.108 ± 0.021
145	615.5 ± 26	0.102 ± 0.004
193	901.7 ± 57	0.112 ± 0.007
286	1143.6 ± 32	0.096 ± 0.002
350	1503.2 ± 81	0.103 ± 0.006
397	1763.7 ± 43	0.107 ± 0.003
	Mean	**0.105 ± 0.006**

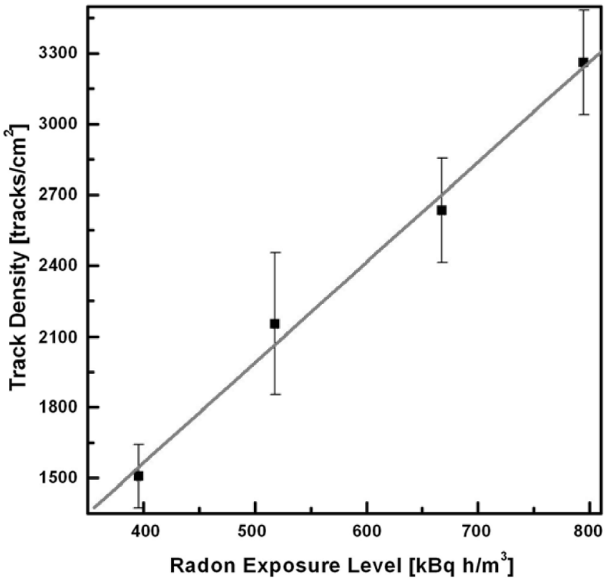

Fig. 10. Result of Linearity Test at Higher Exposure Levels

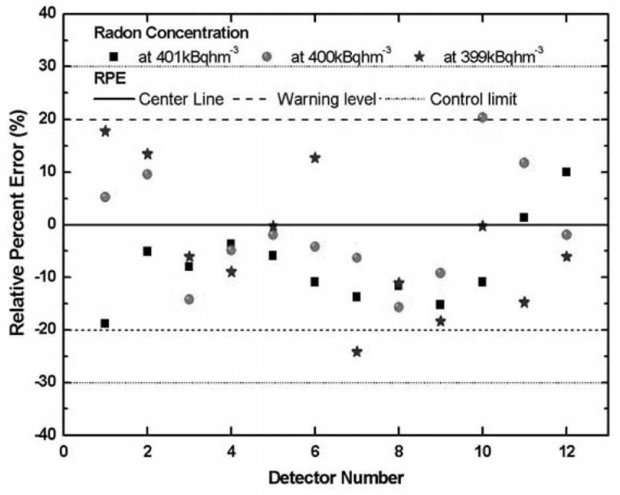

Fig. 11. 101 Result of Intercomparison of Radon Detectors

relative radon concentration normalized over the radon concentration by AlphaGUARD is shown in Figure 11. The radon concentration measured by the radon cup is in an acceptable agreement, within 20 %, with the value obtained by AlphaGUARD. There are many potential sources of error for this result, such as slight changes of etching conditions, different batches of the radon cup, and problems of proper recognition of the etched tracks by the automatic image analyzer [19].

3. CONCLUSION

For large-scale surveys of radon levels in the living environment, a measurement tool should be low cost, easy to distribute, readily available, and simple to evaluate. The proposed environmental radon monitoring system, composed of a radon cup, a chemical etching system, and a track counting system was constructed. The developed radon cup is a cylindrical chamber with a radius of 2.2cm and a height of 3.2 cm in combination with a CR-39 detector. The optimized etching condition for the CR-39 exposed in a radon environment turned out to be a 6 N NaOH solution at 70 ºC over a 7hour period. The bulk etch rate under the optimized condition was 1.14 ± 0.03 μm h^{-1}. The images of tracks were observed by the track counting system, which consisted of an optical microscope, a CCD camera, and an image processor. The calibration factor of this system was obtained to be 0.105 ± 0.006 tracks cm^{-2} per Bq m^{-3} d. The radon concentration measured by the developed radon cup was in acceptable agreement with the values obtained by another device at intercomparison experiment.

The proposed radon monitoring system can be used to investigate nationwide radon levels to estimate the annual effective dose to the public by radon. The data on the survey using this system will be used as the baseline data to decide the action level for radon in Korea. Finally, this system will contribute effectively for the reduction of lung cancer caused by radon.

REFERENCES

[1] UNSCEAR, *Sources, Effects and Risks of Ionizing Radiations*, United Nations Scientific Committee on the Effects of Atomic Radiation UNSCEAR 2000 Report to the General Assembly, with Scientific Annexes. United Nations, New York(1988).

[2] J.K. Lee, *Radiological risk associated with naturally occurring radioactive nuclides in drinking water and protection standards*, Consumer Affairs Institute, Report on “Debate on the Hazardousness of Radioactive Elements in Drinking Water”, 39-49(1998).

[3] J.K. Lee, *Issues on the radon and uranium content in water of wells in district of Daejeon*, Consumer Affairs Institute, Report on “Debate on the Hazardousness of Radioactive Elements in Drinking Water”, 86-94(1998).

[4] J.K. Lee, *Concerns on elevated radon concentrations in air at some subway stations in Seoul*, Activity Report of the Radon Radiation Research Society in Korea (1997.8.1-1998. 10.31), 301-309(1998).

[5] NRC, *Health Effects of Exposure to Radon*, National Research Council, Biological Effects of Ionizing Radiation (BEIR) VI Report(1999).

[6] B.L. Cohen, C.A. Stone and C.A. Schilken, “Indoor Radon Maps of the United States,” *Health Phys*., Vol 66, No. 2, 201-205(1994).

[7] NRPB, *Radon Atlas of England*, National Radiological Protection Board, NRPB-R290(1996).

[8] G. Jonsson, “Indoor 222Rn measurements in Sweden with the solid-state nuclear track detector technique,” *Health Phys*., Vol. 54, No. 3, 271-281(1988).

[9] H. Mellander and A. Enflo, “The Alpha Track Method Used in the Swedish Radon Epidemiological Etudy,” *Proc. of 5th Inter. Symposium on the Natural Radiation Environment*, 22-28(1991).

[10] S.A. Durrani and R. Ilic, *Radon Measurements by Etched Track Detectors*, World Scientific(1997).

[11] M. Urban and E. Piesch, “Low level Environmental Radon Dosimetry with a Passive Track Etch Detector Device,” *Radiat. Protect. Dosim*., Vol. 1, No. 2, 97-109(1981).

[12] H.A. Khan, E. Qureshi and M. Tufail, “Passive Dosimetry of Radon and its Daughters Using Solid State Nuclear Track Detectors,” *Radiat. Protect. Dosim*. Vol. 46, No. 3, 149-170(1993).

[13] G. Espinosa, V.M. Castano and J.I. Golzarri, “Permeability of Some Materials to Radon,” *Nucl. Tracks Radiat. Meas*., Vol. 19, No. 1-4, 307-308(1991).

[14] P.F. Green, A.G. Ramli, S.A.R. Al-Najjar, F. Abu-Jarad and S.A. Durrani, “A Study of Bulk-etch Rates and Track-etch Rates in CR-39,” *Nucl. Instrum. Methods*, 203, 551-559(1982).

[15] J. Miles, *The NRPB Radon Environmental Chamber and its use in International Comparisons*, Proc. of Int. Workshop on Radon Monitoring in Radioprotection, Environmental Radioactivity and Earth Sciences, ICTP. Trieste, 98-109 (1989).

[16] KINS, *Assessment of Radiation Risk for the Korean Population and Nationwide Indoor Radon Survey in Korea*, Korea Institute of Nuclear Safety, KINS/GR-246(2001).

[17] J. Lee, K.H. Ahn, H.S. Chai, T.S. Park, “Development of radon calibration chamber at KRISS,” *Appl. Radiat. Isot*., Vol. 61, 237-241(2004).

[18] Genitron Instruments, Heerstrabe 149, Germany. Model PQ2000PRPO, Portable Raon Monitor, December 1998.

[19] EPA, *National Radon Proficiency Program - Guidance on Quality Assurance*, United States Environmental Protection Agency, EPA 402-R-95-012(1997).