

STUDY OF THE CALIBRATION OF THE MEDICAL PHYSICS DEPARTMENT-RADON DOSIMETER IN A RADON FACILITY

<u>D.Nikolopoulos</u>, A.Louizi, D. Papadimitriou, C. Proukakis, Medical Physics Department, University of Athens, Greece

1. INTRODUCTION

Several techniques have been developed to measure radon indoors. The use of a Solid State Nuclear Track Detector (SSNTD) closed in a cup, has turned out to be the most appropriate for long term measurements. The Medical Physics Department of the Athens University is carrying out radon measurements in dwellings, apartments, outdoor air and mines since (Louizi et al., 1996a). For this purpose a simple device, the so called Medical Physics Department (MPD) radon dosimeter, has been constructed, which measures the radon concentration averaged over a long period of time. In the present paper the calibration technique introduced and the results of the calibration of the MPD dosimeter are reported.

2. DESCRIPTION OF THE MPD DOSIMETER

The dosimeter consists of a 2x2 cm CR-39 polymeric film obtained from Pershore Mouldings LTD, fixed in the base of a cylindrical cup of radius R=1.5 cm and height H=5 cm. Radon gas penetrates into the cup through a 3mm hole on the cup's lid and a filter just below the lid, which prevents radon daughters and other aerosols from entering. The radon in the cup decays through its radioactive chain and some of the alpha particles emitted hit CR-39 and leave damaged trails (latent tracks).

The dosimeters are exposed to radon for a period of time. After the exposure the CR-39 is removed and etched preferentially in 6M NaOH at a constant temperature of 70° C for 13.5 h. The etchand is produced from NaOH crystals and its strength is periodically checked by titration. The constant temperature is maintained by etching on a thermostatically controlled water bath.

The detectors are counted under an optical microscope or with the use of an Image Analysis System (IAS). The system consists of a CCD camera, an optical microscope, a TV monitor and a computer. The computer is using a radon track analysis program (GIPSRAD, Image House) for the analysis and counting of the radon tracks. The whole system is calibrated in the Medical Physics Department.

3. RADON EXPOSURE FACILITY

For calibration purposes a radon exposure facility has been constructed by the Medical Physics Department of the Athens University and the Nuclear Engineering Section of the National Technical University of Athens. The facility has been described elsewere (Louizi et al., 1996b). Radon is generated from a dry radium (226Ra) source (Pylon 2000A). The concentration of the radon gas inside the chamber is measured by a portable radon monitor (Alpha Guard, Genitron Ltd.). Radon progeny radiation is continuously monitored by a method introduced by Louizi et al. (1996b).

In order radon emanation from the source to start, a nut on the top of the source housing has to be removed. This manual way of starting the emanation would impose severe inconvenience since one has to open the container's door in order to handle the source. To overcome this inconvenience the source with the nut removed was enclosed in a small metal drum (source container). By means of a small flow circulator which lies outside the 1m³ steel clad and a valve arrangement which can be remotely controlled, the radon gas produced inside the source container, is put into circulation and carried in the chamber via suitable piping. The circulation process may be repeated if necessary. The variation of the radon concentration inside the chamber depends upon the strength of the radon source and the iteration frequency of the circulation process. By properly controlling the radon gas flow a stable concentration may be achieved.

4. RESPONSE OF THE MPD DOSIMETER

The SSNTD inside the cup registers every alpha particle that can reach its surface except those that have energies above the detector energy threshold. This determines a sampling volume, around the detector surface inside which physically acceptable trajectories may be generated. The distribution of number of tracks in an area S is given by

$$n = \int_{0}^{\pi} c_{j} dt \int_{U(S)}^{\pi} dU \int_{S}^{\pi} dS \cos z / (4\pi r^{2})$$
 (1)

where c is the activity concentration inside the cup (supposedly uniform), T is the exposure time, U(S) the sampling volume, r the length of trajectory from emission position down to detector's surface and z is the angle with the normal at the incidence point. Since registrable alpha trajectories are generated not only from radon but also from radon daughters, ²¹⁸Po and ²¹⁴Po respectively, it is expected that n would be the sum of n_j (j=0,1,4) where n_j is the track distribution due to each member of the decay chain separately. If R_a is the range of alpha particles in air then

$$U(S) = \frac{2}{3}\pi R_a^3 + \pi R_a^2 S^{\frac{1}{2}} + R_a S$$
 (2)

in the case of a square shape detector of area S (Gil et al., 1995). Since each member in the group has a different range R_a the sampling volumes will also be different. Thus the index j will also separate among the sampling volumes $U_j(S)$. In this paper j=0 for radon, j=1 for ^{218}Po and j=4 for ^{214}Po .

The filter prevents the radon daughters from infiltrating or exfiltrating and thus all the daughter nuclides produced will either deposite on cup's surfaces (plate-out) or suspend in the cups' air. All radon daughters deposited on surfaces produce nuclides that remain on the surface, with the exception of ²¹⁸Po that has a small probability of recoil (Knutson 1988). On the other hand according to Hashemi-Nezhad and Peak (1993) almost all ²¹⁸Po isotopes are expected to plate out on the surfaces for a cup of radius ≤ 4cm and height ≤ 10cm. This leads to the conclusion that for the MPD dosimeter (radius=1.5cm, heigth=5cm) almost all the daughters will be surface deposited and thus the cup's atmosphere will be almost radon free. This is certified also by other investigators for a geometry similar to that of the MPD dosimeter (Hazhemi-Neshad and Peak, 1993; Urban, 1984). Four hours after the beginning of exposure, the alpha emitting daughters will be in radioactive equilibrium with radon. Since both polonium isotopes are surface deposited, the total activity of the walls of the MPD dosimeter, expressed in terms of volume equivalent activity concentration (Bq m-3) of the alpha emitting daughters, will be almost twice the value of radon concentration.

Radon alphas may be emitted from any position inside the sampling volume for radon. The alpha particles from 218 Po and 214 Po decay are due to isotopes deposited on the detector surface and on the cup's walls. According to the position of emission with regard to the detector an alpha particle of initial energy E_j (E_0 =5.49 MeV, E_1 =6.00 MeV, E_4 =7.69 MeV) will have an incidence energy on the detector surface ranging from 0 to E_j . The dip angle will vary according to the angle and the point of the emission. The critical angle I_c of etchability for CR-39 is strongly related to the incidence energy according to the relationship $I_c(E)$ =3.8+61.9 E -22.27 E_j (Barillon et a.,1995). Thus, according to the position and the kind of the emission, alpha particles with various incidence energies and angles will reach the detector, a fraction of which will be registrable. Taking into account the random nature of the alpha emissions and assuming a homogeneous distribution of each alpha emitting nuclide within the cup, it is concluded that the above fraction will be constant for each nuclide independent of the concentration. This leads to the conclusion that registration volumes $U_{R_j}(S)$ inside the MPD dosimeter will be constant for each one of ^{222}Rn , ^{218}Po and ^{214}Po .

The track shape depends upon the dip angle and the etching conditions. The track area depends on the incidence energy of the alpha (Handler et al., 1991; Handler and Paulo, 1994; Barillon et al., 1995). Above 2 MeV the area of the tracks is inversely proportional to the energy while below a different behaviour exists (Handler et al., 1991). The tracks corresponding to energies of 7 MeV are dark. For E<7MeV the tracks are brighter as the alpha energy is lower, so that, for E≈0, the tracks present almost the same gray level as the surface of CR-39 (Handler and Paulo, 1994). By fixing the etching conditions a fixed track shape and area distribution inside MPD dosimeter is expected. This has been verified experimentally (Nikolopoulos et al., 1997)

If the counting of tracks is done without rejection criteria, then the track density growth rate in the CR-39 mounted on the base of the MPD dosimeter will originate from four sources:

(1) Decay of radon in the cup's air. According to Fleischer and Morgo-Campero (1978) this is given by

$$\rho_1 = (C_{cup}/4) R_0 \cos^2 l_c$$
 (3)

where C_{cup} is the concentration of radon inside the cup (supposedly uniform), R_0 the range of alpha particles from radon and I_c is the critical angle of etchability of tracks originating from radon. Since I_c is energy dependent a mean value for all incidence energies should be adopted. This value is 49° (Barillon et al., 1995)

(2) Tracks from alpha particles emitted at the surface of the detector., i.e. as 218 Po and 214 Po. The registration efficiency for each nuclide is $(1-\sin I_c)/2$ and since radon is in equilibrium with its daughters for long exposure times as that involved in integrated measurements $C_{218}=C_{214}=C_{cup}$ and therefore the growth rate will be

$$\rho_2 = C_{\text{cup}} (V/S) [1 - (\sin I_{c1} + \sin I_{c2})/2] (4)$$

where V and S refer to the volume and surface area inside the cup (Hashemi-Nezhad and Peak, 1993). Ic1,Ic2 are the critical angles of etchability of full energy alphas of 218Po and 214Po respectively. Again mean values should be adopted which are 39° for 218Po and 27° for 214Po (Barillon et al., 1995).

- (3) Tracks due to alpha particles emitted by Po isotopes plated-out on the walls that are capable of reaching the detector. The growth rate from this source is ρ_3 and since the concentration of the deposited nuclides is proportional to the concentration of radon inside the cup, ρ_3 is proportional to C_0 .
- (4) Track density, ρ_4 , due to radon daughters plated-out on the detector surface decaying after the removal from the cup.

The overall accumulated track density in the CR-39 detector in the MPD dosimeter when exposed to radon for a period of time t will be:

$$\rho = (\rho_1 + \rho_2 + \rho_3)t + \rho_4$$
 (5)

Since ρ_1 , ρ_2 , ρ_3 are proportional to C_{cup} the above equation can be written as

$$\rho = K C_{cup} t + \rho_4 \qquad (6)$$

where K is a constant of unit length. K depends upon the geometry of the cup and is thus called the overall efficiency of the dosimeter.

The above result is also valid in the case of a non constant C_{cup} with the substitution of C_{cup} t with $\int_{a}^{\infty} C_{cup}(t) dt$. Therefore,

$$\rho = K \int_0^\infty C_{\text{cup}}(t) dt + \rho_4 (7)$$

In radon dosimetry involving measurements at low radon concentrations, long exposure times are involved whereas in calibration experiments short exposure times at high concentrations. In each case ρ_4 is very small compared to the integral $K \int C_{cup}$ (t)dt. and thus can be neglected.

Therefore

$$\rho = K \int_0^\infty C_{\text{cup}}(t) dt \quad (8)$$

i.e. the track density of the CR-39 detector is proportional to the accumulated exposure to the radon in the cup. Many investigators have shown that the track density increases linearly with the accumulated radon exposure of the environment in which the dosimeter is put (e.g. Frank and Benton, 1977; Jojo et al., 1994; Fleischer et a., 1980; Khan et al., 1990; Subba Rammou et al., 1988) certifying in that way that for a period of exposure $\int_0^{t_0} C_{\text{cup}}(t) dt$ is proportional to $\int_0^{t_0} C_{\text{env}}(t) dt$ where C_{env} is the concentration of the environment in which the dosimeter is put and $\int_0^{t_0} C_{\text{env}}(t) dt$ the accumulated exposure of the dosimeter.

Experiments were carried out in order to evaluate the correlation between the measured track density and the accumulated radon exposure of the MPD dosimeters. The experiments have been reported elsewere (Nikolopoulos,1996). The total results have shown a linearity with an adjusted correlation coefficient r^2 of 0.996 for optical measurements and 0.998 for measurements with the IAS. The above values indicate a very close relationship for both types of measurements. The mean calibration factor obtained equals 4.62 ± 0.33 (RSD=4.6%) for optical measurements and 2.99 ± 0.14 (RSD=7.1%) for IAS measurements. The differences between the mean calibration factors in each exposure indicate an underestimation of the recorded track densities as these are counted by the IAS. This underestimation arises mainly due to track rejection criteria employed by the IAS for the counting of tracks. A second source of the underestimation of the IAS arises from the fact that overlapping tracks are not easily counted separately. For this reason the IAS cannot measure correctly over 1100 KBq m-3 h.

The lower limit of detection is the 2 σ value of the background tracks. This can be interpreted as the accumulated radon concentration in which the relative standard deviation equals 50%. The background track density of the CR-39 sheets kept in Medical Physics Department lie below 10 tracks cm⁻². Considering the maximum background track density of 10 tracks cm⁻² the lower detection limit for the measurement of a 0.36 cm² field, is 6574 Bq m⁻³h. Thus the MPD dosimeter will measure a low concentration of 9.1 Bq m⁻³ with a one month exposure or a high concentration of 400 Bq m⁻³ with a 16 day exposure.

5.REFERENCES

- Barillon R., Fromm M. and Chambaudet A. (1995) Variation of the critical angle of alpha particles in CR39: implications for radon dosimetry. *Radiat. Meas.* Nos 1-4, 631-634.
- Fleischer R. L., Giard W. R., Morgo-Campero A., Turner L.G., Alter H.W., and Ginrich J.E., Dosimetry of environmental Radon: Methods and Theory for Low-Dose, Integrated Measurements. *Health Phys.* (39), 957-962.
- Fleischer R. L. and Morgo-Campero A. (1978) Mapping of integrated radon emanation for detection of long-distance migration of gases within the earth: techniques and principles. J. Geophys. Res. 83, 3539-3549
- Frank A. L. and Benton E. V. (1977) Radon Dosimetry using Plastic Nuclear Track Detectors *Nucl. Track Detect.* I(3/4), 149-179.
- Gil L.R., Marques A. and Rivera A. (1995) SSNTD calibration and equilibrium factor of radon progeny. *Radiat. Meas.* Vol.24, No. 2, 107-109.
- Handler N. J., Iunes P. J., Osorio A. A. and Paulo S. R. (1991) Relationship between track size and energy for alpha particles in CR-39. *Nucl. Tracks Radiat. Meas.*, 19(1-4), 313-317.
- Handler N. J. C. and Paulo S. R. (1994) Indoor radon daughter contamination monitoring: the absolute efficiency of CR-39 taking into account the plateout effect and environmental conditions. *Radiat. Prot. Dosim.*, Vol.51, No.4., 283-296.
- Hashemi-Nezhad S.R. and Peak L.S. (1993) Measurements of plate-out rates of ²²²Rn daughters in laboratory conditions: implications for radon dosimetry. *Nucl. Tracks Radiat. Meas.* Vol.21, No.2, 225-234.
- Islam G. S. and Haque A.K.F (1994) Measurement of mixed radon and thoron daughter concentrations using alpha and beta activities filtered from air. Radiat. Meas. 23(4) 737-742.
- Jojo P. J., Khan A. J., Tyagi R. K., Ramachandran T.V., Subba Ramu M.C. and Prassad R. (1994) Interlaboratory Calibration of track-etch detectors for the measurement of radon and radon daugther levels. *Radiat. Meas.* (23)4, 715-724.
- Khan A. J., Varshney A. K., Prasad R., Tyagi R. K. and Ramanchandran T. V. (1990) Calibration of CR-39 plastic track detector for the measurement of radon and its daughters in dwellings. *Nucl. Tracks Radiat. Meas.* 17, 497-502
- Knutson E. O. (1988) Modeling indoor concentrations of radon's decay products. In: Radon and its decay products in indoor air. (eds Nazaroff W.W. and Nero A. V. Jr.) pp.162-163, John Wiley and Sons. New York.
- Louizi A., Koukouliou V. and Proukakis C., (1996a) A last report of radon concentration studies in Greek dwellings. In: *Proceedings of the first Mediterranean Congress on Radiation Protection* (eds Giakoumakis E., Anagnostakis M. J. and Petropoulos N.P.) pp.398-399. Press of the National Technical University of Athens, Athens.
- Louizi A., Proukakis C., Petropoulos N., Hinis E., Simopoulos S. and Leonidou P. (1996b) Mini Radon House: A prototype container for the measurement of radon exhalation from building materials and structural modules. In: Proceedings of the first Mediterranean Congress on Radiation Protection (eds Giakoumakis E., Anagnostakis M. J. and Petropoulos N.P.) pp.252-268. Press of the National Technical University of Athens, Athens.
- Nikolopoulos D., Louizi A., Petropoulos N., Koukouliou V., Simopoulos S., Proukakis C., (1997) Parametric study of the calibration of track-etch detectors in a radon facility, Rad. Meas. (in press)
- Nikolopoulos D. (1996) Calibration of track-etch dosemeters constructed by Medical Physics Department-University of Athens, Conference on Radon and Greek dwellings, 6 June 1996
- Subba Ramu M.C., Murleedharan T. S. and Ramanchadran T.V. (1988) Calibration of a solid state nuclear track detector for the measurement of indoor levels of radon and its daughters. Sci. Total Environ. 73, 245-255.
- Tommasino L. (1990) Radon Monitor by Alpha Track Detection Nucl. Tracks Radiat. Meas. (17)1, 43-48.
- Urban M. (1984) Calculated and measured sensitivity of the Karlsruhe radon diffusion chamber using makrofol track etch detectors and different ECE techniques. *Nucl. Tracks Radiat. Meas.* Vol 1-4, Nos 1-4, 429-432.
- Ward W. J., Fleischer R.L. and Morgo-Campero A. (1977) Barrier technique for separate measurements of radon isotopes. Rev. Sci. Instrum. 48, 1440, 1441