Qatar Univ. Sci. J. (1994), 14 (2): 234-237

MEASUREMENTS OF RADON PERMEABILITY THROUGH SOME MEMBRANES

BY

W. ARAFA*
Department of Physics, University of Qatar. Doha, Qatar

قياس نفاذية غاز الرادون خلال بعض الأغشية

وفـاء محمود عرفـه

تمت دراسة إنتشار غاز الرادون - ٢٢٢ خلال أنواع مختلفة من الأغشية بواسطة طريقة تعتمد على الإتزان الإشعاعي وباستخدام كاشف سيليكون نو قدرة فصل ١٤ كيلوالكترون فولت لجسيمات ألفا الصادرة من عنصر الأمريسيوم - ٢٤١ . وقد تم تعيين ثابت النفاذية لعينات من الميلار المغطى بالألومنيوم وكذلك لعينات من عديد الاثيلين ونترات السيليلوز والبيسفينول - أ عديد الكربونات ، بالاضافة إلى عينات من ورق الحائط المتوفر تجارياً . وتمت مقارنة النتائج بتلك النتائج المنشورة سابقاً ، كما تمت دراسة تأثير سمك أغشية البيسفينول - أ عديد الكربونات على ثابت نفاذية غاز الرادون .

Key Words: Measurements of Radon, Radon permeability, Membranes.

ABSTRACT

The diffusion of ^{222}Rn through different types of membranes has been studied by an equilibrium method using a Si surface barrier a-detector with a resolution of 14 keV FWHM for ^{241}Am. The permeability constant for Aluminized mylar, Aluminized polycarbonate, Polyethylene, Cellulose nitrate, Bisphenol-A polycarbonate and different types of wall paper have been evaluated and compared with previous data. The effect of membrane thickness on the permeability constant of Bisphenol-A polycarbonate is discussed.

INTRODUCTION

Interest for radon monitoring in air, water and soil is increasing rapidly. In addition to radon hazards for public health,measurements of its concentration is of great interest in mining as well as earth sciences. In order to estimate the concentration of the radon gas, solid state nuclear track detectors (SSNTD) were used . These detectors are sensitive for all alpha particles emitted from the two isotopes of the gas ^{222}Rn,^{220}Rn (known as thoron), from their decay products and from any other alpha emitters that may exist near the detector.In some applications special membranes are used to separate ^{222}Rn from ^{220}Rn. Such membranes delay the diffusion of the gas (Tanner, 1964) and consequently a large percentage of the long lived ^{222}Rn will pass through the membrane while a large percentage of the short lived ^{220}Rn will decay during its diffusion.Some other applications need a membrane to filter only the decay products of the gas, while other membranes are needed to stop completely the gas (Mclaughlin, 1979; Ward *et al.*, 1977; Wojcik, 1991).

The diffusion of the gas through membranes is function of the permeability constant of the membrane material. In general, the permeability constants for different materials are not widely available in the literature. However,published values (Abdel Fattah *et al.*, 1986; Bigu, 1986; Giridhar *et al.*, 1982; Pohl-Ruling *et al.*, 1980; Ramachandran *et al.*, 1987; Wojcik, 1991) for a given commercially available material vary considerably due to the difference in manufacturing procedures and physical properties of the same material. Consequently,we have found it interesting to measure the permeability constant of some commercially available materials and some samples of wall paper. Wall paper of low permeability constant might be suitable to reduce radon concentration in dwellings.

In our measurements we used surface barrier α- detector instead of the commonly used solid state nuclear track detectors or Zinc Sulphide scintillator.The advantage of the surface barrier detectors lies in its good alpha energy resolution. By these detectors, we can detect radon by counting mainly the 5.49 MeV α line following its decay. All other α- particles are considerably eliminated. This advantage cannot be fulfilled in previous similar measurements using SSNTD or ZnS scintillator.

PHYSICAL CONSIDERATION

The method used in the present work is similar to that described by Ramachandran *et al.* (1987). It is based on placing a radon source (2.33 MBq of ^{226}Ra) in a fixed large volume V_1 as shown in (Fig.1). Two small volumes V_2 ($V_2 << V_1$) are included in V_1 . One of these two volumes is open to V_1 while the second is separated from V_1 by a membrane.

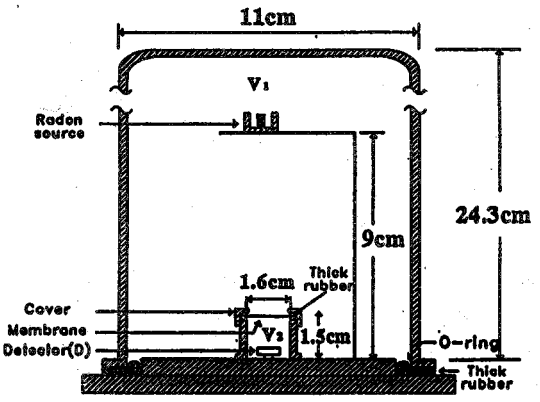

Fig. 1: Schematic diagram of the alpha chamber.

After equilibrium, the alpha counts in the first small volume (N_1) will be taken as a measure for radon concentration in the large volume V_1 while alpha counts in the second small volume (N_2) will be taken as a measure for radon concentration after diffusion through the membrane. In the work of Ramachandran both N_1 and N_2 were measured simultaneously by using two SSNTD.

In the present work, we have used only one small volume V_2 and one surface barrier Si detector to detect alpha rays. N_1 and N_2 were measured in two steps. In the first step, N_1 was measured without membrane after reaching equilibrium. IN the second step N_2 was measured with the membrane after reaching the equilibrium. The time to achieve equilibrium was measured experimentally by following the counting rate as function of time. It was found to be 7 days. According to Ramachandran *et al.* (1987) the variations of N_1 and N_2 is given by

$$\frac{dN_1}{dt} = -\lambda N_1 + a - \frac{KA}{\delta V_1}(N_1 - N_2) \qquad (1)$$

$$\frac{dN_2}{dt} = -\lambda N_2 + \frac{KA}{\delta V_2}(N_1 - N_2) \qquad (2)$$

where

a is the production rate (λN_1) of radon ($cm^{-2}s^{-1}$)

λ is the radon decay constant (s^{-1}),

K is the permeability of the membrane (cm^2s^{-1}),

δ is the thickness of the membrane (cm) and A is its effective area (cm^2).

So for different boundary conditions and with the assumption that t > mean-life of radon,we can define R as the ratio between the concentration of the radioactive gas in volume V_2 with a membrane present and the concentration without any membrane present.

$$R = \frac{N_2}{N_1} = \frac{KA(V_1+V_2)}{KA(V_1+V_2)+V_1V_2\lambda\delta} \qquad (3)$$

and the permeability constant is

$$K = \frac{V_1V_2\lambda\delta}{A(V_1+V_2)}\left(\frac{N_2}{N_1-N_2}\right) \qquad (4)$$

EXPERIMENTAL

The experimental set-up is shown in Fig.2 . It consists of :

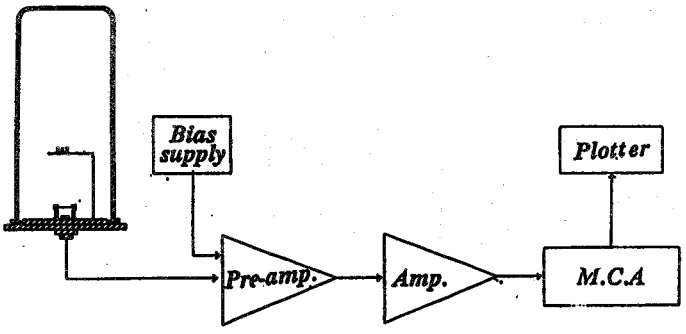

Fig.2 : Block diagram of experimental set-up.

An ORTEC a-chamber model 807* acts as the radon chamber with V_1=2305 cm^3. A small chamber of volume V_2= 3 cm^3 was fixed around the detector in such a way that a membrane can be fixed on its top. Special care has been taken to ensure that radon can diffuse only into V_2 through the membrane. This has been done by ensuring a good sealing around the small chamber especially the bottom and by putting two thick rubber rings up and down the membrane. The ^{226}Ra source was placed not facing the detector in such a way that the detector cannot sense the 4.79 MeV line for ^{226}Ra. The detector is an ORTEC partially depleted surface barrier detector type BA 14-50-100* with active area 50 mm^2 and depletion depth of 100 μm. Its resolution is 14 keV FWHM for the alpha line 5.486 MeV (^{241}Am). The detector is biased by +75 volts using an ORTEC model 428 bias supply*. Pulses from the detector were amplified by an ORTEC spectroscopy amplifier model 571* after passing through an ORTEC charge sensitive preamplifier model 142*. The amplified pulses were analyzed by a 2048 channels analyzer Nucleus card** mounted on an AT IBM personal computer***

Fig.3 shows the α spectrum from the ^{226}Ra source when placed facing the detector in the detection volume V_2. The spectrum has four well resolved α lines of energies 4.79 , 5.49 , 6.00 and 7.69 MeV. These lines belong to ^{226}Ra, ^{222}Rn, ^{218}Po and ^{214}Po respectively. When the source was placed not facing the detector at distance of 9 cm from the detector, the spectrum obtained (Fig.4) has two small peaks at energies ≈ 6 & 7.7 MeV in addition to

scattered counts down to energy equal zero. Since the ^{226}Ra α line cannot reach the detector, the two small peaks should correspond to ^{218}Po and ^{214}Po . The scattered particles counted in the energy range from zero to about 5.5 MeV belong to all α particles emitted by ^{222}Rn gas plus portions of scattered particles from its daughters. In all our measurements, we have selected a counting region of interest ranging from zero to 5.5 MeV to minimize counting particles from radon daughters. It should be noted that the detector noise near the zero energy level was cut by the threshold potentiometer in the analyses system.

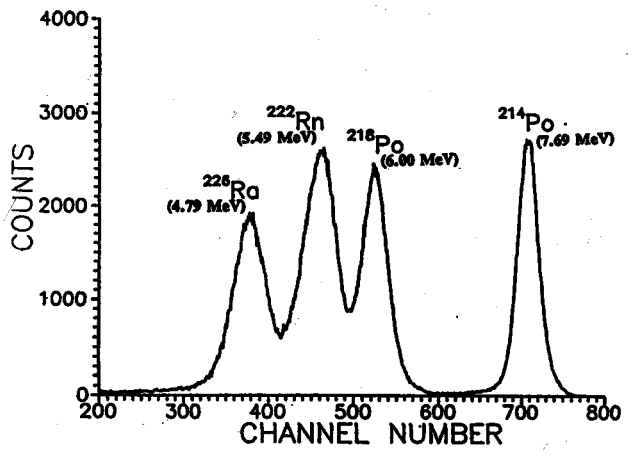

Fig.3: Alpha spectrum from ^{226}Ra and its daughters (the source facing the detector).

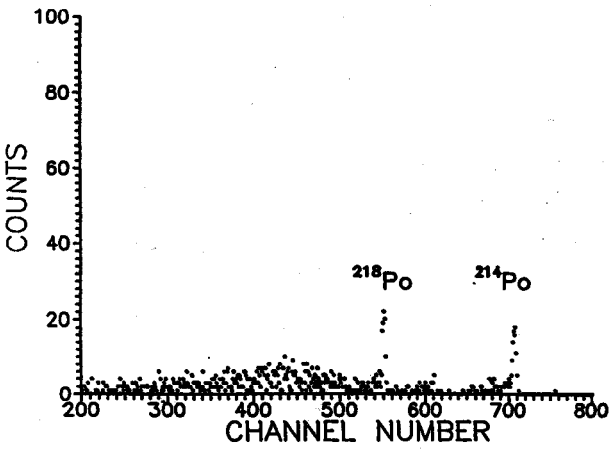

Fig.4: Alpha spectrum from ^{222}Rn and its daughters (the source not facing the detector).

RESULTS AND DISCUSSION

In the present work five different commercially available materials in addition to some samples of commonly used wall paper were studied. These materials are : Aluminized mylar, Aluminized polycarbonate, Polyethylene, Cellulose nitrate and Bisphenol-A polycarbonate . The measurement of each sample were repeated several times in order to obtain an average value. The obtained results are summarized in Table 1 and includes the membrane or wall paper thickness, the ratio N_2/N_1,and the estimated permeability constant. In all these results, an estimated error of about 17% was found. In all our measurements, the sample effective area was kept constant at 2 cm^2. From the obtained results we can summarized the following:

Table 1

Permeability constant of ^{222}Rn in some materials.

Material Studied	Sample Thickness(μm)	$R=N_2/N_1$	Permeability Constant(m^2s^{-1})
Aluminized mylar	35	0.72	28.5×10^{-13}
Aluminized-polycarbonate	20	0.38	3.8×10^{-13}
Polyethylene	40	0.49	1.87×10^{-13}
Cellulose nitrate (LR 115)	12	0.29	1.5×10^{-12}
Makrofol	12	0.33	1.8×10^{-13}
Makrofol	10	0.39	2.0×10^{-13}
Makrofol	8	0.43	1.9×10^{-13}
Makrofol	5	0.60	2.4×10^{-13}
Wall paper [a]	236	0.62	1.2×10^{-11}
wall paper [a]	226	0.80	2.9×10^{-11}
wall paper [b]	351	0.79	4.2×10^{-11}

a) polyamide (laminated plastic material)
b) cellulosic material (impregnated fabric cotton)

1. The permeability constant of Polyethylene membrane of thickness 40 μm was found to be 1.87×10^{-13} m^2s^{-1}. This is in the same order of magnitude of the value 3.35×10^{-13} m^2s^{-1} previously reported by Ramachandran *et al.* (1987) for membrane of thickness 25.1μm.

2. The permeability constants of Aluminized polycarbonate of thickness 20 μm and Aluminized mylar of thickness 35 μm were found to be 3.8×10^{-13} and 28.5×10^{-13} m^2s^{-1}, respectively. These values cannot be compared with previous data available for polycarbonate and Mylar membranes. However, permeability constants for Polycarbonate membrane (Ramachandran *et al.*, 1987; Abdel-Fattah *et al.*, 1986) varies between 3.8×10^{-13} for thickness 25.3 μm to 0.55×10^{-16} m^2s^{-1} for thickness 14.8 μm. For Mylar, Ramachandran *et al.* (1987) found $K=8.37 \times 10^{-14}$ m^2s^{-1} for thickness 17.6 μm while Abdel-Fattah and Somogy (1986) reported a value of 0.3×10^{-16} m^2s^{-1} for thickness of 12 μm.

3. The permeability constant for Cellulose nitrate membrane with thickness 12 μm was found to be 1.5×10^{-12} m^2s^{-1}. This value is about 10 times smaller than the value of 1.25×10^{-11} m^2s^{-1} previously reported by Ramachandran *et al.* (1987) for cellulose nitrate material of thickness 14.7 μm. This difference may be attributed to the difference in the additional chemical composition and cristallinity of polymer materials.

4. The values of K for wall paper samples are in the order of 10^{-11} m^2s^{-1}. This indicates that these types of wall papers can

not be used efficiently to reduce radon concentration in dwellings.

5. The result obtained for Bisphenol-A polycarbonate membrane with thickness varies between 5 to 12 μm are shown in Table.1. Fig.5 shows the relation between the membrane thickness δ and the ratio N_2/N_1-N_2 . It is clear that N_2/N_1-N_2 is inversely proportion to δ in the thickness range from 8-12 μm. This may indicate that the permeability constant K is constant for this range. At smaller thickness ,the permeability constant K increases sharply. However more studies are necessary to establish this relation . For δ less than 8 μm, the permeability constant increases to ≈ 2.4 $x10^{-13}$ m^2s^{-1} at thickness of 5 μm. At small thickness 2 μm, the value of N_2/N_1 was found to be ≈1, so the permeability constant cannot be determined for such small thickness.

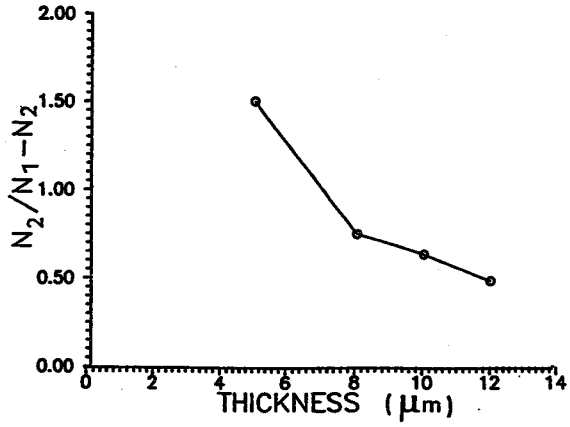

Fig.5: The variation of N_2/N_1-N_2 versus the thickness δ for Bisphenol-A polycarbonate (Makrofol) membrane.

ACKNOWLEDGEMENTS

The author is thankful to Prof. Dr. L. Al-Houty, Chairman of the physics department for her useful advice, QATAR university for offering the use of experimental facilities. The author wishes to express his gratitude to Prof.Dr. H. Abou Leila for very helpful discussions concerning interpretation of the results.

Footnote

* On leave from Ain Shams University,Faculty of women ,Cairo-Egypt.

+ EG&G ORTEC,100 Midland Road,Oak Ridge,Tennessee 37830 U.S.A .

++ The Nucleus,INC. 761 Emory Valley Road.Oak Ridge. TN 37831-2561 .

+++International Business Machines Corporation P.O.Box 1328-W Boca Raton,Florida 33429-1328 .

REFERENCES

[1] **Abdel-Fattah, H.** and **G. Somogy, 1986.** Determination of radon and thoron permeability through some plastics by track technique. Nucl. tracks 12: 697-700 .

[2] **Bigu, J., 1986.** An automated multisensor apparatus for comparative membrane radon and thoron permeability studied. Nucl. Instr. and Methods A251: 366- 373 .

[3] **Giridhar, J., M. Raghavayya,** and **N. Padmanabham, 1982.** Radon permeability of some membranes. Health Phys. 42: 723-725 .

[4] **Mclaughlin, J. P., 1979.** A technique for measuring the relative exhalation rates of radon and thoron from building materials. Presented at the E.E.C./S.C.P.R.I. seminar on the Radiological Burden of Man from Natural Radioactivity in the Countries of the European Community; Vesinet, France;4-6 December .

[5] **Pohl-Ruling, J., F. Steinhausler** and **E. Pohl, 1980.** Investigation on the suitability of various materials as ^{222}Rn diffusion barriers. Health Phys. 39: 299-301 .

[6] **Ramachandran, T.V., B.Y. Lalit** and **U. C. Mishra,** 1987. Measurement of radon permeability through some membranes. Nucl. Tracks Radiat. Meas. 13: 81-84 .

[7] **Tanner, A. B., 1964.** Natural radiation environment. In: Adams, J.A.S.; Lowder, W.L. Chicago: University of Chicago Press.

[8] **Ward, W. J., III, R. L. Fleischer,** and **A. Margo-Campero, 1977.** Barrier techniques for separate measurement of radon isotopes. Rev. Sci. Instrum. 48: 1440-1441.

[9] **Wojcik, M., 1991.** Measurement of radon diffusion and solubility constants in membranes. Nucl. Instr. and Methods B61: 8-11.