SNO-STR-93-052

PERMEABILITY PROPERTIES OF URYLON-453 (3.26mm) TO ^{222}Rn

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ABSTRACT

The permeability and diffusion coefficient of a material (Urylon-453, thickness 3.26mm) to ^{222}Rn ha been measured using two different techniques. The first technique made use of radioactivity (^{222}Rn) measurements under steady-state (i.e., radioactive equilibrium) conditions, whereas the second technique required continuous radioactivity monitoring, i.e., radioactivity versus time measurements. The experimental apparatus consisted of twin chambers separated by the material of interest. Radioactivity measurements were conducted using two identical counting systems. The permeability (k) of the material was measured to be 3.44×10^{-10} (cm^2/s) and the diffusion coefficient (D) was found to be 2.34×10^{-8} (cm^2/s).

INTRODUCTION

A desirable feature of certain materials is their ability to separate the components of a gas mixture by diffusion processes. A variety of natural and artificial materials commonly available on the market have properties that make them ideally suitable for a variety of applications for industrial, chemical, medical, biological, pharmaceutical, commercial and research purposes. In addition to a number of natural chemical compounds, a wide range of artificially produced materials are potentially useful in a variety of separation applications. Equally important in many applications is the very low, or even total lack of permeability characteristics to a particular type of gas. Materials with very low permeability charcacteristics to some particular fluid or group of fluids are usually referred to as 'barriers'. Clearly, the basic difference between a 'membrane' and a 'barrier' resides in their degree of permeability to a given fluid or group of fluids.

In this report the permeability characteristics of Urylon-453 to ^{222}Rn are investigated. Studies have been conducted using a 0.326cm sample. The material is potentially useful as a ^{222}Rn barrier to reduce the ^{222}Rn flux density from mine walls.

EXPERIMENTAL APPARATUS

The experimental apparatus used is shown in Figure 1. It consists of two cylindrical chambers of identical geometry and physical dimensions. Each chamber has an open end which is terminated with a flange of suitable dimensions. The two chambers are assembled by matching the two open ends and tightening the flanges. The material of interest is placed (sandwiched) between the flanges of the two chambers. This geometrical configuration will be referred to as the twin-chamber method. Each chamber is provided with two ports (inlet and outlet at opposite ends of the chamber) to allow the radioactive gas, e.g., ^{222}Rn to flow through the chamber.

A source of ^{222}Rn (i.e., ^{226}Ra) is placed in a leak-free enclosure provided with inlet and outlet ports to allow the ^{222}Rn formed by the decay of ^{226}Ra to be directed to one of the chambers, i.e., the 'source' chamber. The other chamber (i.e., the 'test' chamber) is used to monitor the amount of ^{222}Rn that penetrates the permeable material sandwiched between the two chambers. Each chamber is connected to identical sampling trains and counting systems. Each sampling train and counting system consists of a sampling pump, a filter/filter holder system of the in-line type (to remove ^{222}Rn progeny before entering the counting system), a scintillation cell/PMT assembly and associated nuclear scaler, as well as the necessary tubing and on/off gas valves. Both sampling/counting systems operate in a recirculation mode. Some of characteristics of the twin-chamber method used here are given below:

Volume of each chamber : $V = 3551$ cm^3
Surface area of open end of each chamber : $S = 118.8$ cm^2
^{226}Ra 'source' strength : $SE = 2.2 \times 10^3$ kBq
Scalers/PMT : Model AB-5 (Pylon Electronics Development, Ottawa)
Scintillation cells : ~ 150 cm^3 (in-house made)

THEORETICAL BACKGROUND

Two theoretical approaches and experimental methods have been used in the measurements reported here, namely: the steady-state method, and the dynamic method.

STEADY-STATE METHOD

In this method, measurements under steady-state conditions, i.e., radioactive equilibrium, are used to calculate the permeability,k, of the material. It can be shown that for the geometry used here, k is given by:

$$k = (R/1-R)(\lambda\delta V/2S) \qquad (1)$$

where,

λ is the radioactive decay constant of ^{222}Rn, i.e., 2.1×10^{-6} /s
δ is the thickness of the sample ($\delta = 0.326$ cm)
V is the volume of either chamber ($V = 3551$ cm^3)
S is the surface area of the material ($S = 118.8$ cm^2)

The variable R is defined as the ratio of ^{222}Rn concentration, [^{222}Rn], in the 'test' chamber relative to the 'source' chamber ($R \leq 1$).

However, in order for Equation 1 to hold for a radioactive gas such as ^{222}Rn, the following two conditions must be met:

$$\theta = \delta^2/6D \ll 0.693/\lambda$$

and,

$$DL = (D/\lambda)^{1/2} \gg \delta \qquad (2)$$

where θ is the so-called time-lag (see below), DL is the diffusion length, and D is the diffusion coefficient of the material. The variables D and k are related by the expression:

$$k = Ds \qquad (3)$$

where s is the solubility of the material.

It is not difficult to see that the above conditions are not met (see below) for the material tested. This is due to the excessive thickness of the sample, i.e., $\delta = 0.326$cm. Hence the results reported here can only be considered to be rough estimates with undetermined errors associated with them.

DYNAMIC METHOD

In this method , [^{222}Rn] measurements versus time are conducted. From the slope (linear region) of the R-curve (where R has been defined above), the solubility of the sample, s, can be calculated:

$$s = (\Delta R/\Delta t)(\delta V/SD) \qquad (4)$$

The diffusion coefficient, D, can be calculated from θ (see Equation 2), the time-lag, which can be obtained graphically from the intersection of the [^{222}Rn] versus time curve (see Figure 3). However, in order for $\theta = \delta^2/6D$ to apply some conditions must be met.

Again, because of the value for δ (0.326cm) these conditions are not met, and hence, the results are not conclusive and are affected by an undetermined error. In any case, in order to determine D with sufficient accuracy, samples as thin as possible should be used. It should also be noted that the expression for θ is only approximate. However, a discussion on this topic is beyond the scope of this report (see ref. 1).

EXPERIMENTAL RESULTS AND DISCUSSION

Some of the experimental data are shown in Figures 2 to 4. Figure 2 shows the ^{222}Rn concentration, [^{222}Rn], versus time in the 'source' chamber. Figure 3 shows the growth of [^{222}Rn] versus time in the second chamber, i.e., ^{222}Rn diffusing through the material. This quantity is shown as a continuous line. Figure 4 shows the ratio R (defined above) versus time. From Figures 2 and 3, or alternatively, from Figure 4, the value for k can be calculated using Equation 1. The steady-state R value is obtained by averaging the ^{222}Rn concentration during the last 24 hours of the experiment (see Figure 4 for the graph of R versus time). With $R = 3.36 \times 10^{-5}$, one obtains:

$$\begin{aligned} k &= (3.36\mathrm{x}10^{-5})/(1-3.36\mathrm{x}10^{-5}) \times (2.1\mathrm{x}10^{-6}/\mathrm{s} \times 0.326\mathrm{cm} \times 3551\mathrm{cm}^3)/(2 \times 118.8\mathrm{cm}^2) \\ &= 3.44 \times 10^{-10}\ (\mathrm{cm}^2/\mathrm{s}) \end{aligned}$$

Furthermore, from Figure 3, the time-lag can be estimated by graphical extrapolation. Taking $\theta = 210$ h, the diffusion coefficient can be calculated:

$$\begin{aligned} D &= \delta^2/6\theta = (0.326\mathrm{cm})^2/(6 \times 210\mathrm{h} \times 3600\mathrm{s/h}) \\ &= 2.34 \times 10^{-8}\ (\mathrm{cm}^2/\mathrm{s}) \end{aligned}$$

The solubility, s, can be calculated using Figure 4 and Equation 4, hence,

$$\begin{aligned} s &= (1.8\mathrm{x}10^{-5})/((400\mathrm{h}-210\mathrm{h})\mathrm{x}3600\mathrm{s/h})\mathrm{x}(0.326\mathrm{cm})(3551\mathrm{cm}^3)/(118.8\mathrm{cm}^2\mathrm{x}2.34\mathrm{x}10^{-8}\mathrm{cm}^2/\mathrm{s}) \\ &= 1.10\mathrm{x}10^{-2} \end{aligned}$$

The solubility, s, can be calculated from k and D obtained by the steady-state method and the dynamic method, respectively. From Equation 3 one obtains:

$$\begin{aligned} s &= k/D = 3.44 \times 10^{-10}\ (\mathrm{cm}^2/\mathrm{s}) / 2.34 \times 10^{-8}\ (\mathrm{cm}^2/\mathrm{s}) \\ &= 1.47\mathrm{x}10^{-2} \end{aligned}$$

The two values obtained for solubility agree reasonably well.

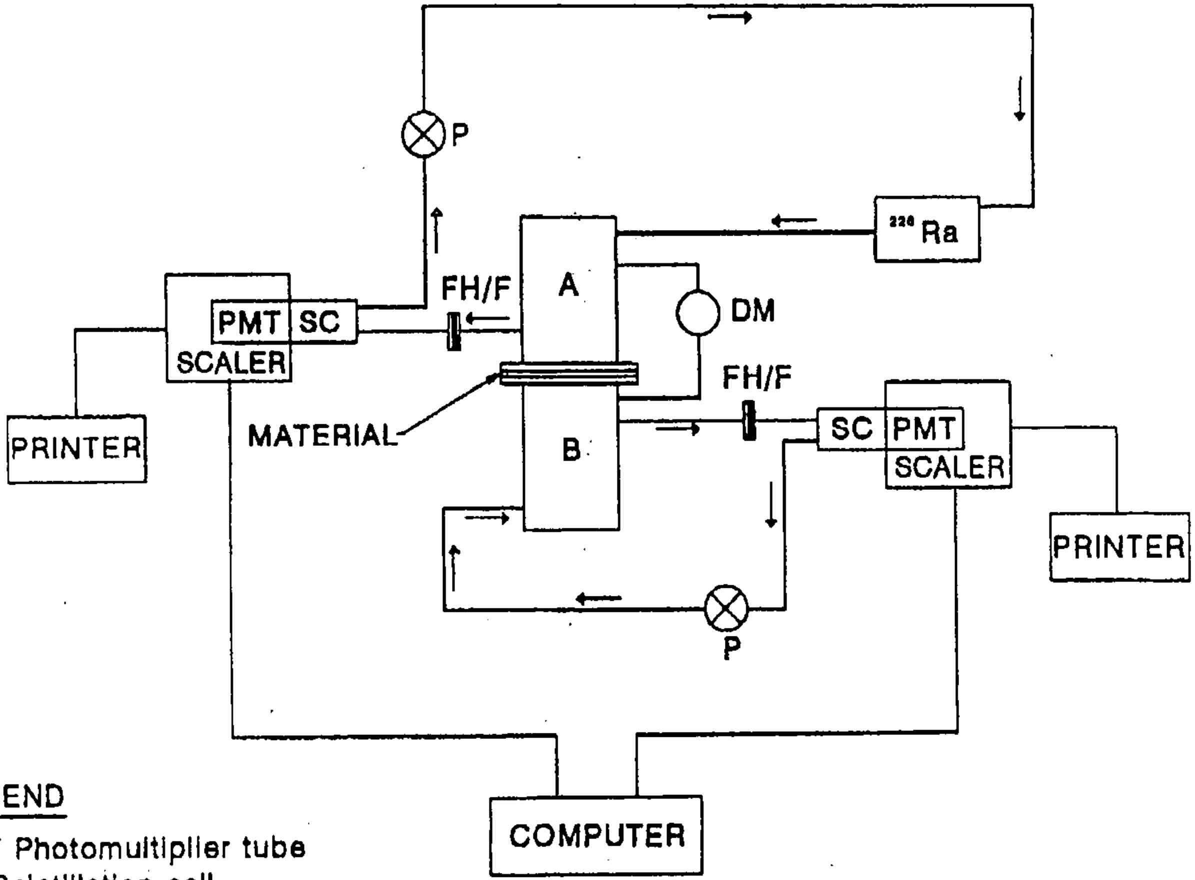
REMARKS AND CONCLUSIONS

The large value for δ precludes some theoretical conditions to be met, and hence, the results given in this report are affected by unknown errors. More accurate measurements of D can be carried out by reducing δ so that the conditions indicated by Equation 2 are met.

REFERENCES

Bigu, J., Hallman, E.D. and Kendrick, L. "Permeability of different materials to radon (^{222}Rn) gas". Sudbury Neutrino Observatory Project Report SNO-STR-91-069, 1991. (See Division Report MRL 91-153(TR), CANMET, Energy, Mines and Resources Canada, 1991.)

Bigu, J., Hallman, E.D. and Patrakka, P. "Permeability Properties of Urylon-453 to ^{222}Rn". Division Report MRL 93-033(TR), CANMET, Energy, Mines and Resources Canada, Ottawa 1993.

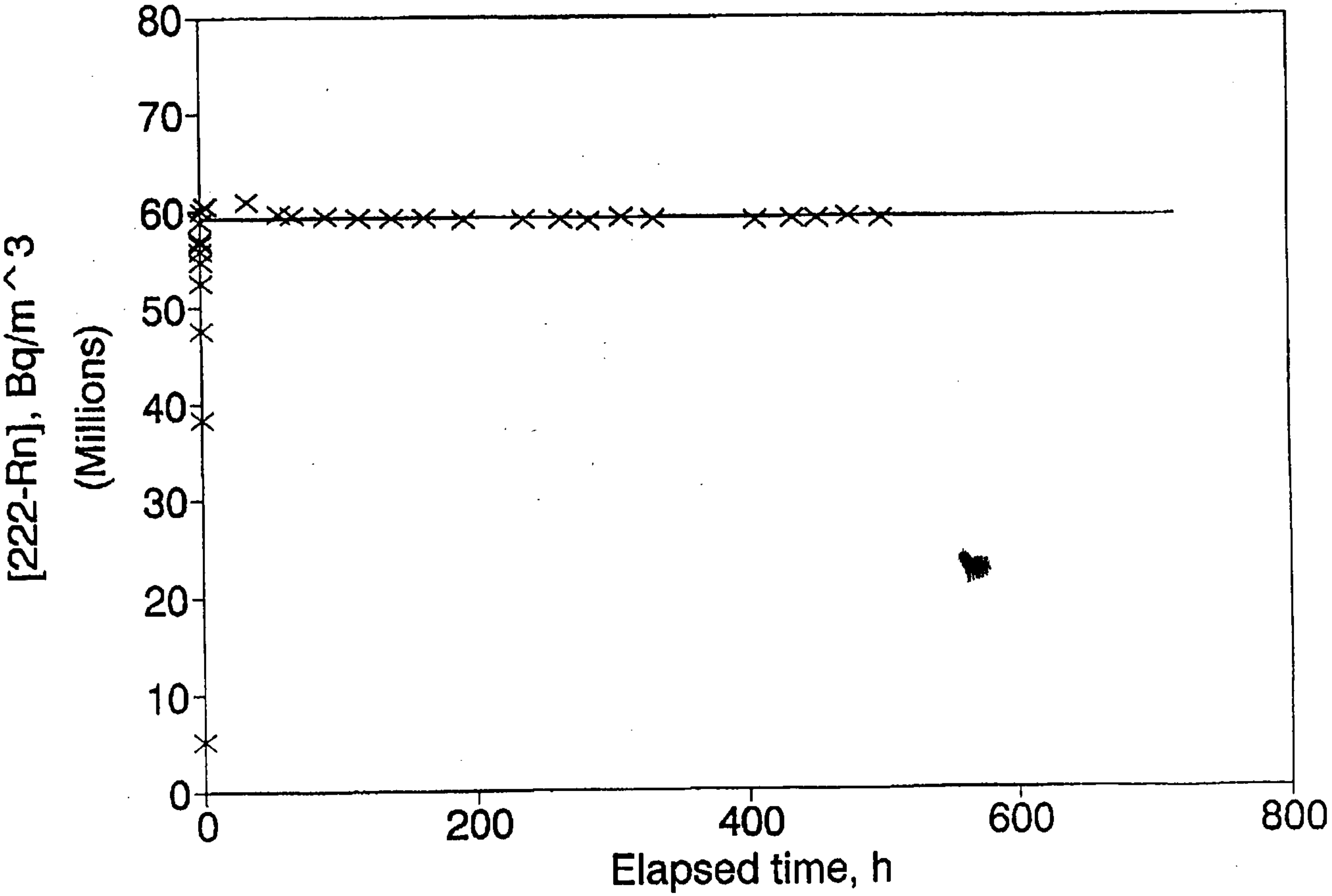


LEGEND

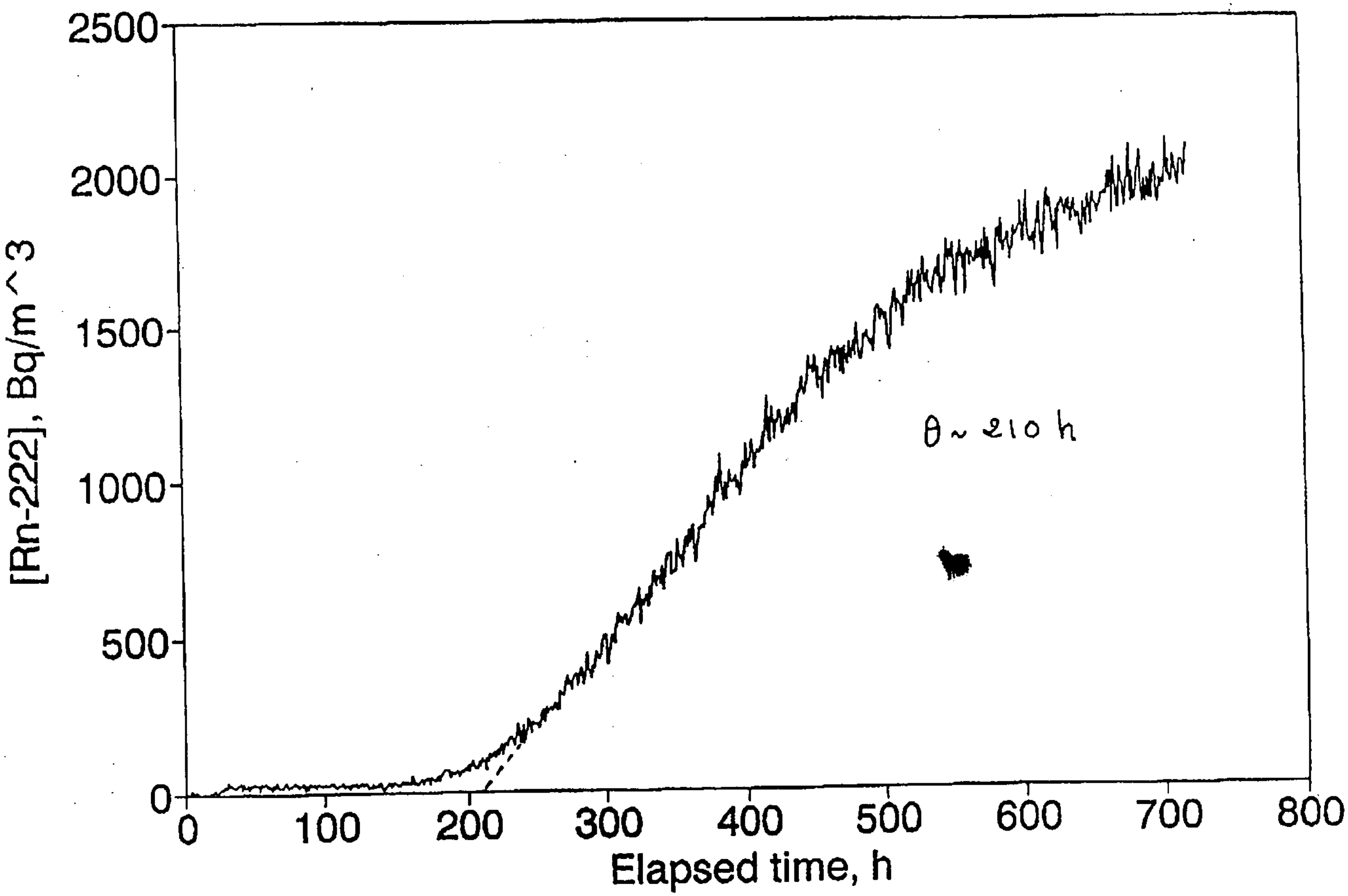
PMT Photomultiplier tube
SC Scintillation cell
P Pump
DM Differential manometer
FH/F Filter holder/filter system
A Upper chamber
B Lower chamber

Fig. 1

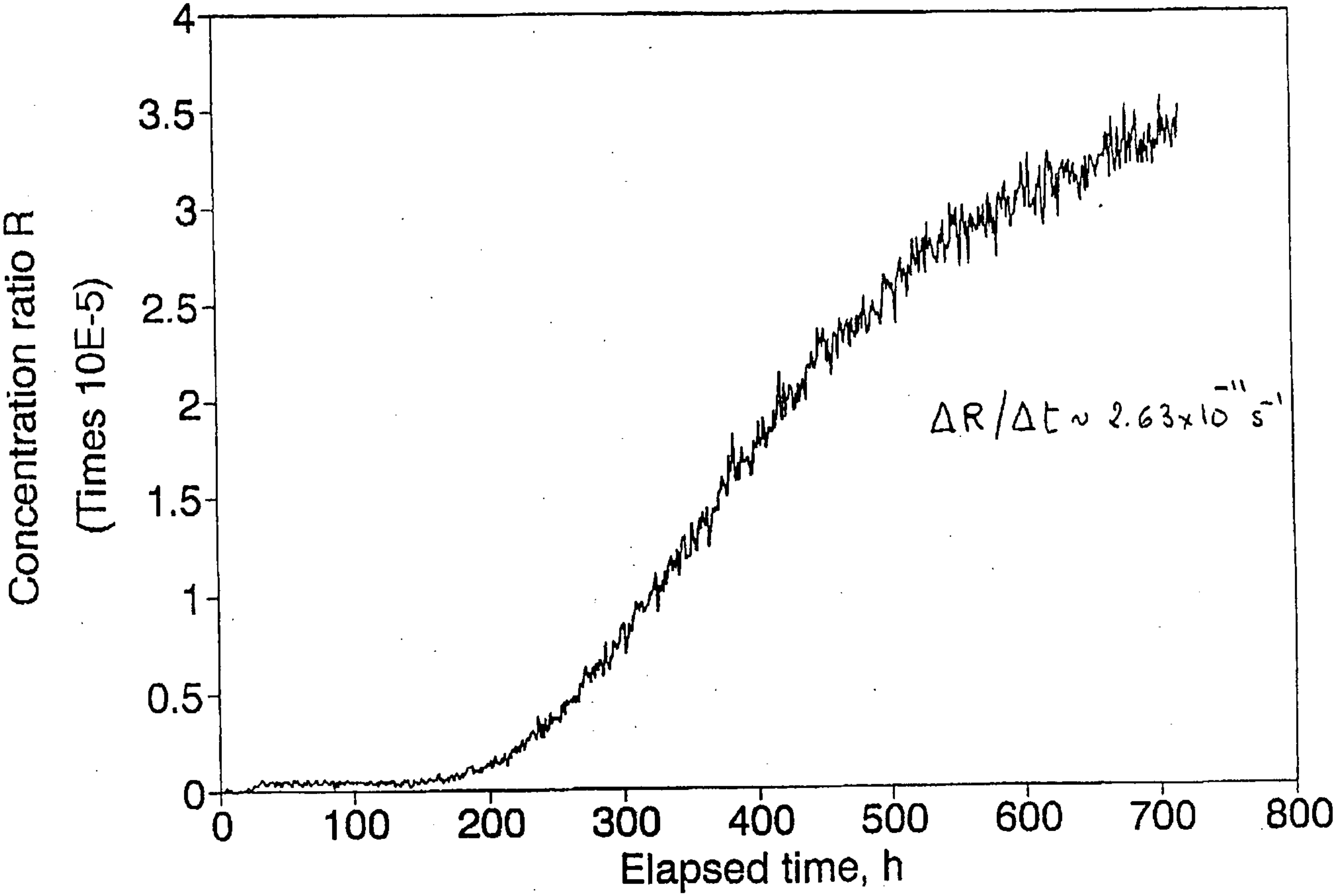
[222-Rn] IN "SOURCE" CHAMBER
Urylon 3.26mm



[Rn-222] IN TEST CHAMBER vs TIME
Urylon 3.26mm



Concentration ratio R vs Time
Urylon 3.26mm



$\Delta R/\Delta t \sim 2.63\times10^{-11}\,s^{-1}$