SNO-STR-93-016

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Division Report MRL 93-033(TR) April 1993

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ABSTRACT

The permeability and diffusion coefficient of a material (urylon-453) to ^{222}Rn has 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 of the order 10^{-11} cm^{2}s^{-1}.

Key words: Permeability; Diffusion; Radon; Radon barriers.

*Research Scientist and Head of Radiation Section, Elliot Lake Laboratory, CANMET, Energy, Mines and Resources Canada, and Adjunct Professor, Laurentian University, Sudbury, Ontario; **Professor, Laurentian University, Sudbury, Ontario; ***Research Technologist, Laurentian University, Sudbury, Ontario.

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 of these materials to a particular type of gas. Materials with very low permeability characteristics 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 thick (~1 cm) 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 here 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 the 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:

$$k = (R/1-R)\ (\lambda \delta V/2S) \tag{1}$$

where, λ is the radioactive decay constant of ^{222}Rn, i.e., 2.1×10^{-6} s^{-1}

δ is the thickness of the sample ($\delta = 1.14$ 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:

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

and,

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

where, θ is the so-called time-lag (see below), 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., $\delta > 1$ cm. Hence, the results reported here can only be considered to be rough estimates with undetermined errors associated to 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:

$$s = \left(\frac{\Delta R}{\Delta t}\right) \frac{\delta V}{SD} \qquad (4)$$

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 (>1 cm) 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. The error associated with the measurements is indicated by the broken lines which define a 'confidence' envelope. 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. From Figure 4, R ~ 0.5 x 10^{-6}, hence:

$$k = \left(\frac{0.5 \times 10^{-6}}{1 - 0.5 \times 10^{-6}}\right) \frac{2.1 \times 10^{-6} \times 1.14 \times 3551}{2 \times 118.8} \sim 1.8 \times 10^{-11}\ cm^2\ s^{-1}$$

Furthermore, from Figure 3, the time-lag can be estimated by graphical extrapolation. The value obtained is rather imprecise but in the range 50-100 h. Taking θ ~50 h, the diffusion coefficient can be calculated:

$$D = \delta^2/6\theta = \frac{(1.14)^2}{6 \times 50 \times 3600} \sim 1.2 \times 10^{-6}\ cm^2 s^{-1}$$

The solubility, s, can be calculated using Figure 4 (see Figure 4A) and Equation 4, hence,

$$s \sim \frac{0.4 \times 10^{-6} \times 1.14 \times 3551}{(240 - 90) \times 3600 \times 118.8 \times 1.2 \times 10^{-6}} \sim 2 \times 10^{-5}$$

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:

$$s = k/D \sim 1.8 \times 10^{-11}/1.2 \times 10^{-6} = 1.5 \times 10^{-5}$$

The two values obtained for the solubility agree reasonably well although they are much smaller than expected. The results obtained for D and s are much larger and smaller, respectively, than anticipated. No experimental flaws have been found in the experimental procedure or data analysis. However, the data are under review to ensure the validity of the results.

REMARKS AND CONCLUSIONS

Although the value calculated for the permeability (k) of the sample is rather small ($\sim 10^{-11}$ cm^2s^{-1}), the diffusion coefficient (D) is not particularly low ($\sim 10^{-7}$ cm^2s^{-1}). The small value obtained for k is attributed mainly to the thickness (δ) of the material. The large value for δ precludes some theoretical conditions to be met, and hence, the results are most likely to be affected by large errors. Furthermore, the accuracy of the measurements is rather limited because of the low signal (count rate), and hence, poor statistics of counting. The signal (S) to noise (N) ratio, i.e., ratio of 'total' [^{222}Rn] to [^{222}Rn] background in the 'diffusion' or 'test' chamber was $S/N \lesssim 2$. Accurate measurements of D can be carried out by reducing δ so that the conditions indicated by Equation 2 are met.

REFERENCE

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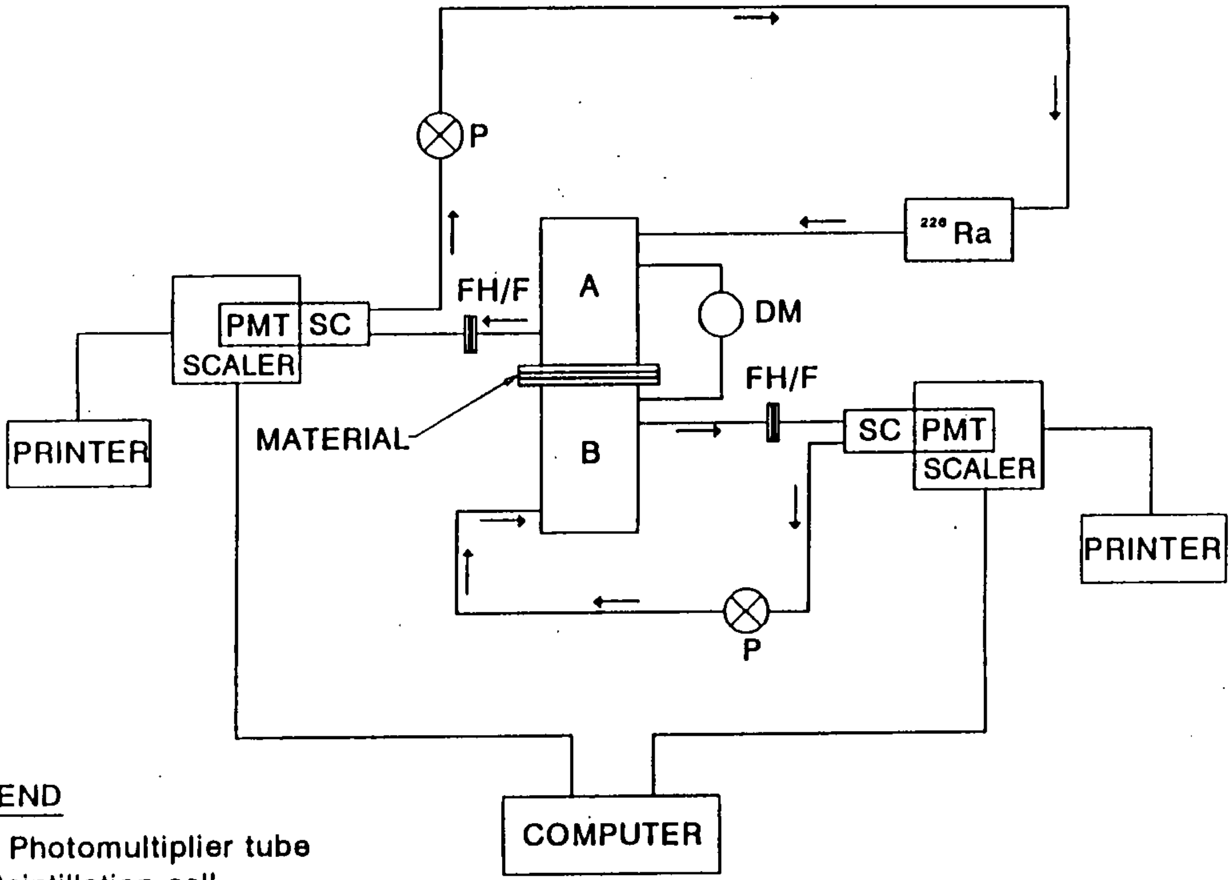

LEGEND

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 - Twin-chamber experimental apparatus for permeability studies.

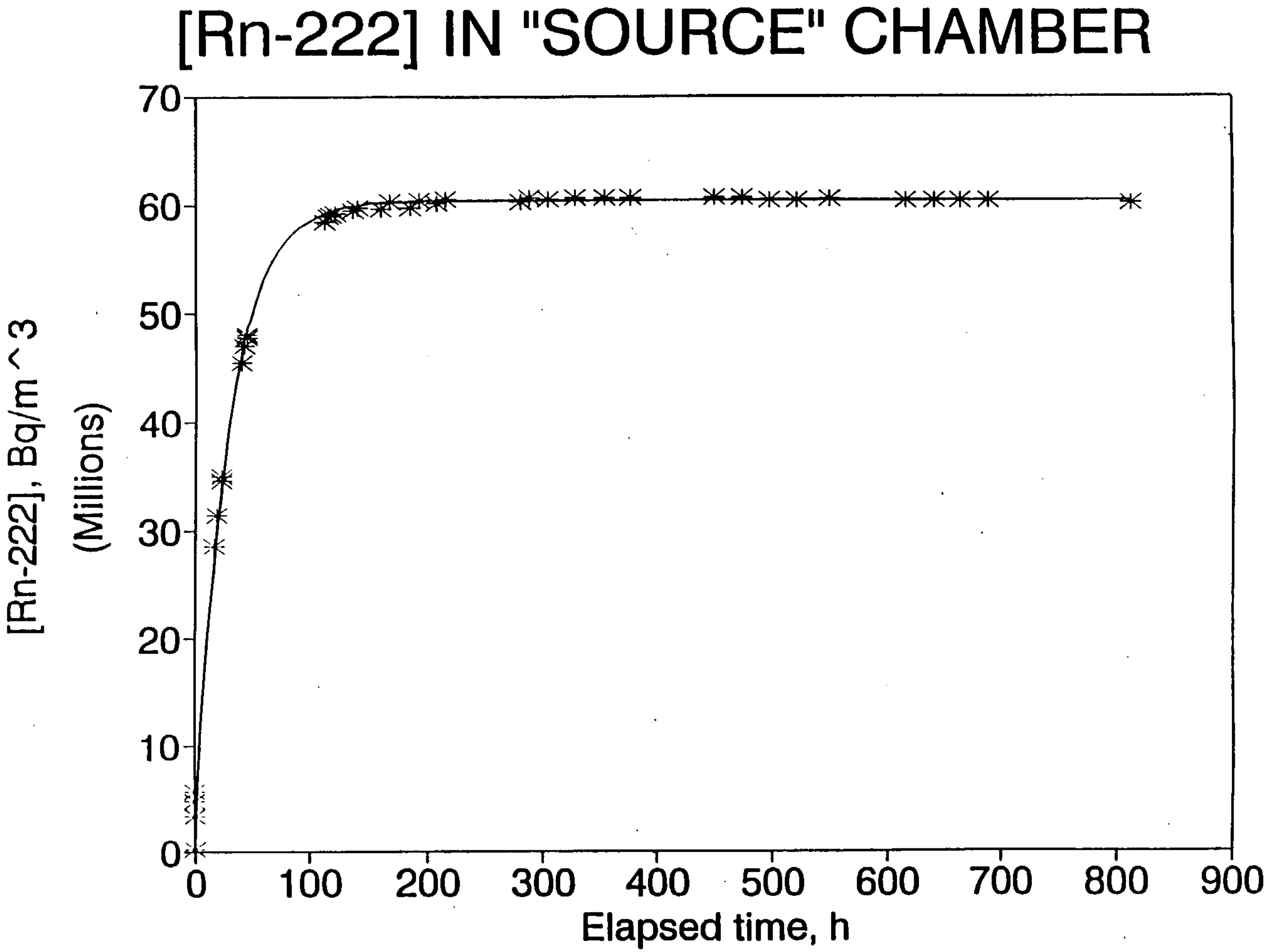

Fig. 2 - In-growth of ^{222}Rn in the source chamber.

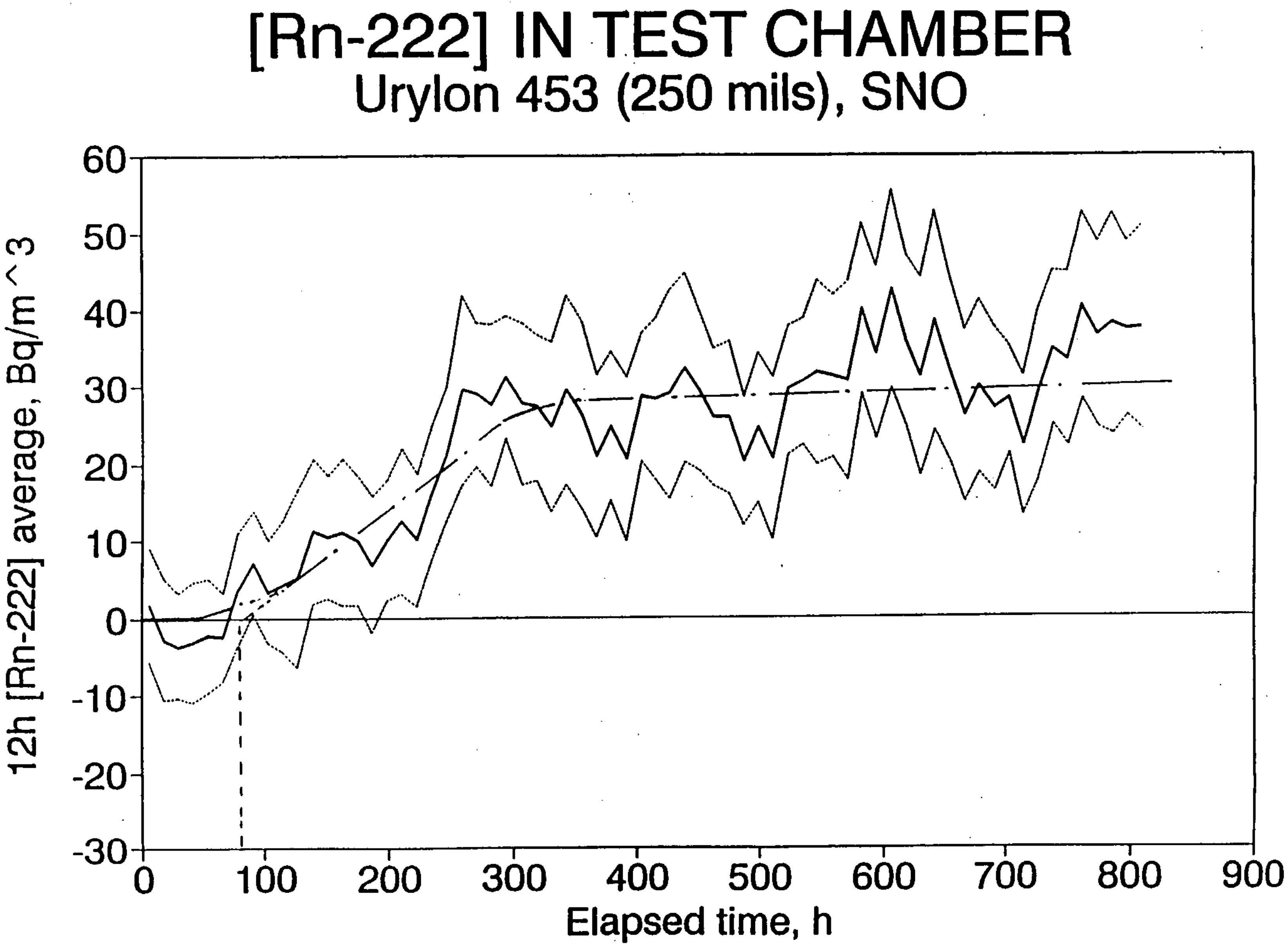

Fig. 3 - Radon gas concentration, [^{222}Rn] in the 'test' chamber versus time (thick continuous line). The short broken lines represent error bands. The 'smooth' broken line represents the theoretical curve. The extrapolated value from the linear portion gives the time-lag, θ.

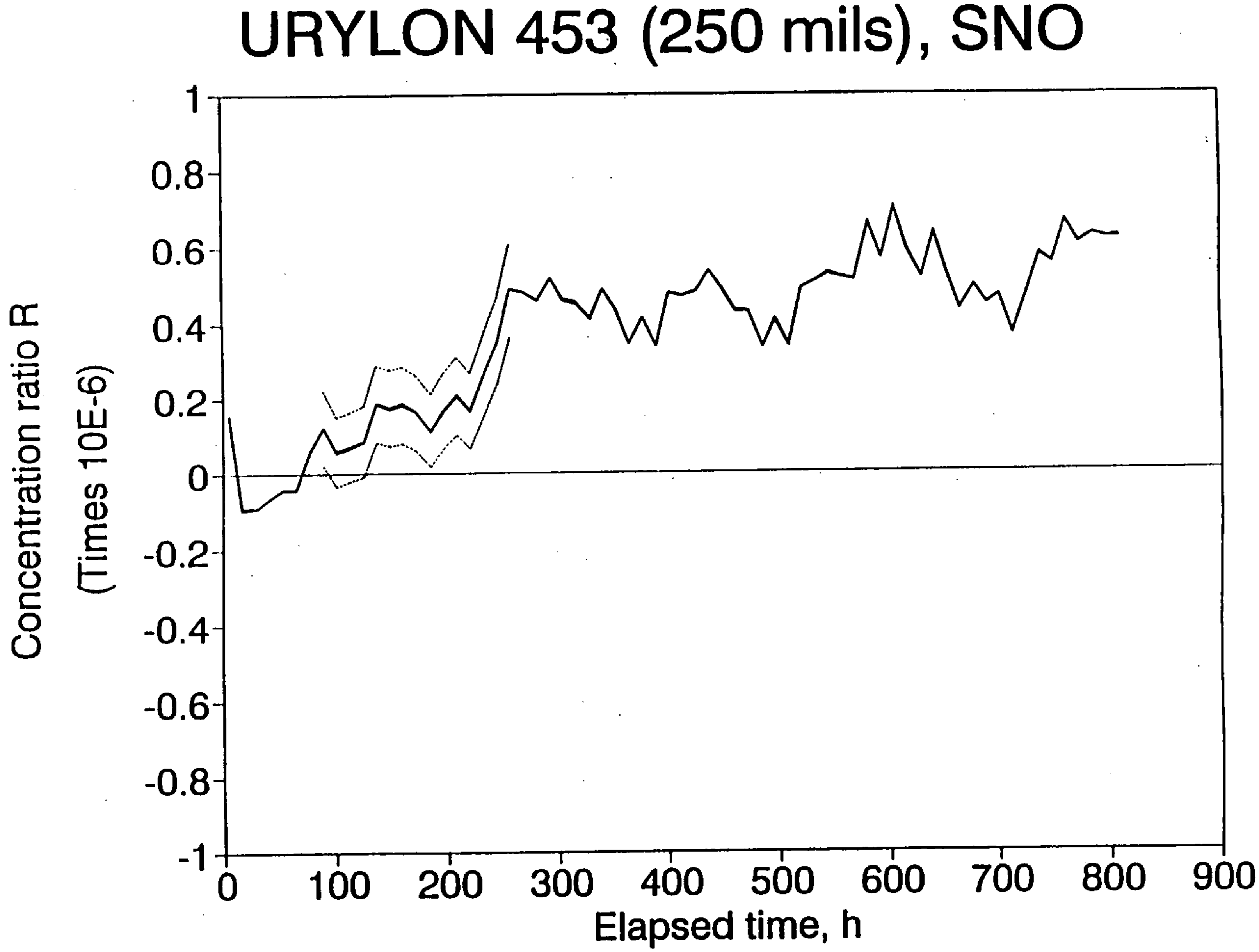

Fig. 4 - Concentration ratio, R, versus time (continuous line). The broken lines represent error bands. The expanded region of interest for the calculation of the solubility is shown in Figure 4A.

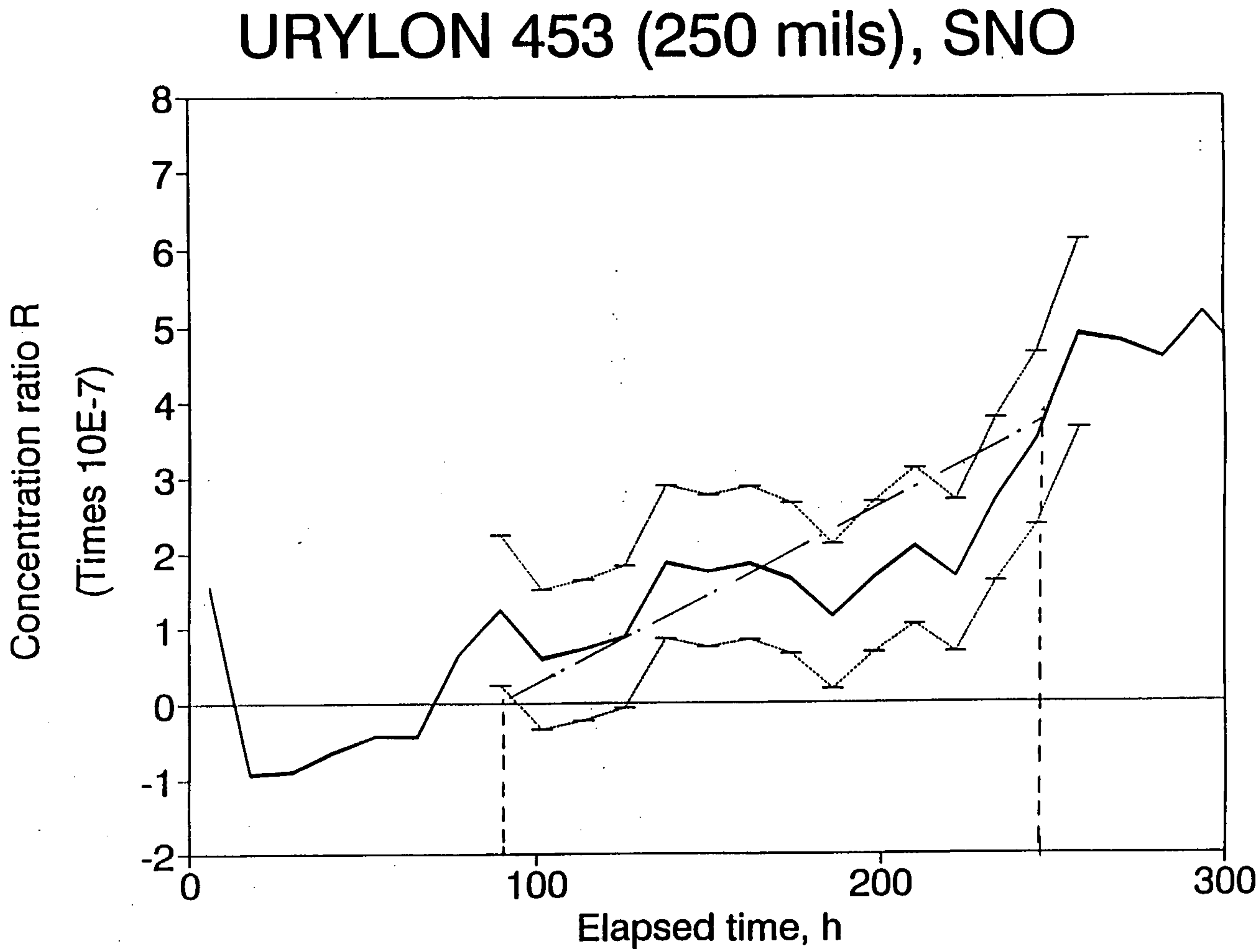

Fig. 4A - Expanded region of Figure 4 used to calculate the solubility, s. Short broken lines represent error bands. The long broken straight line is used to calculate s according to Equation 4.

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PERMEABILITY PROPERTIES OF URYLON 453-85RH AND POLYPROPYLENE

J. Bigu, E.D. Hallman, and P. Patrakka

The permeability properties of urylon 453-85RH and polypropylene were studied by α-particle detection techniques using methods previously described.

1. URYLON 453-85RH

2.2×10^{3}

A plane sheet of the material (thickness: 0.67 cm) was investigated by the twin-chamber method using a ^{226}Ra source of strength 2.2 kBq, and two scintillation cell/photomultiplier tube/nuclear scaler systems. The duration of the experiment was slightly over 6 weeks during which the growth of ^{222}Rn concentration in both chambers, i.e., the ^{226}Ra source chamber and the "permeability" chamber, were continuously and simultaneously monitored. The permeability, k, was calculated according to the simplified expression:

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

where, R is defined as the ratio of ^{222}Rn concentration, [^{222}Rn], in the "permeability" chamber relative to the "source" chamber ($R \leq 1$);

λ is the radioactivity decay constant of ^{222}Rn, i.e., $\lambda = 2.1 \times 10^{-6}\ s^{-1}$;

δ is the thickness of the sample ($\delta = 0.67$ cm);

V is the volume of either chamber ($V = 3551\ cm^{3}$) (both chambers have the same volume);

S is the surface area of the material (urylon), $S = 118.8\ cm^{2}$

The calculated value for R, based on the [^{222}Rn] in both chambers was: $R \sim 0.2 \times 10^{-6}$

The permeability was calculated as:

$$k\ (\text{urylon}) \sim 4.4 \times 10^{-12}\ cm^{2}\ s^{-1}$$

2. POLYPROPYLENE

Four cylindrically shaped (capped at one end) samples (polypropylene tubing) of different wall thicknesses (0.1, 0.3, and 0.6 cm) were studied. The samples were coupled to specially designed detector/electronic circuitry systems, as described elsewhere. The detection systems consisted of a diffused function (DJ) detector housed in a "sensitive" volume terminated in a flange to which the above samples could be "coupled". Results from these samples were compared with the results from two samples to which small holes had been drilled and covered with glass fibre (GF) filter material.

Experiments were conducted in a sealed ^{222}Rn box, 3m^3 in volume in which a 2.2 kBq ^{226}Ra source was located. The ^{222}Rn produced by the decay of the ^{226}Ra source was thoroughly mixed and circulated inside the box by means of a mixing fan. The duration of the experiments was 7 weeks during which continuously monitoring of ^{222}Rn diffusing through the polypropylene and the GF systems was carried out.

The permeability, k, of the material was calculated according to:

$$k = (R / 1-R)\ (\lambda \delta V_2 / S) \qquad (2)$$

where all the symbols in Eq. 2, except V_2, have already been defined (see Eq. 1). The symbol V_2 represents the total "sensitive" volume of the detector system, i.e., the "sensitive" volume of the detector per se plus the inner volume of the cylindrical (tubing) sample. The results obtained are summarized in the table below.

Detector type	Thickness (cm)	k (cm^2s^{-1})
DJ, AL	0.6	4.6×10^{-9}
DJ, AC	0.1	0.3×10^{-9} to 0.7×10^{-9}
DJ, AC	0.3	0.34×10^{-9} to 0.8×10^{-9}
DJ, AC	0.6	0.98×10^{-9} to 2.2×10^{-9}

Note: DJ stands for diffused-junction detector. AL and AC refer to the detector/electronic circuitry arrangement used.

It should be noted that in either case, namely, for Urylon and polypropylene, the more accurate method in which the time-lag, etc can be obtained graphically could not be used reliably because of the poor statistics of counting, i.e., low α-particle counting rates. The simplified method used here, based on the activity measured under radioactive steady-state conditions , is not rigorous because it does not meet an important theoretical requirement. Hence, the method is only approximate but justifiable here because of the difficulties indicated above. For the polypropylene case there is a further source of uncertainty because of the geometry of the sample. I have dealt with this geometry as it were a plane geometry, i.e., by "squashing" the internal surface of the cylinder to a plane "sheet" of the same surface area through which ^{222}Rn would diffuse, hence reducing this case to the plane geometry case discussed in previous reports.

The contribution to the count rate from ^{222}Rn emanating from the sample has not been measured and, hence is "buried" in the count. In addition, and in spite of all the precautions taken, the potential for leaks due to the finish of the polypropylene flanges is a real one.