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DEVELOPMENT OF A TRAP TO MEASURE RADON CONCENTRATION IN A HIGH-FLOW STREAM OF NITROGEN GAS

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1 INTRODUCTION

This report describes work to make and to characterize a trap that can measure the Rn content of the SNO cover gas system. The requirement on the Rn purity of the cover gas is set by the solubility of Rn in water. The Ostwald coefficient, defined as the ratio of the concentration per unit volume of Rn dissolved in heavy water to the concentration per unit volume of Rn in the gas above the heavy water under equilibrium conditions, is equal to 0.385 at 10 C and 0.277 at 20 C [see, e.g., 'Solubility Data Series', vol. 2, pp. 227-235 (1979)]. If it is required that the dissolved Rn contribute no more than 10% to the total allowed upper limit of 10^{-14} g U/g D_2O, then the Rn content of 10 C cover gas must be less than 0.17 Rn atoms/liter. To obtain a sufficient number of Rn atoms for clear detection above Lucas cell background, say 100 atoms, one must thus flow a volume of about 600 liters of gas, and trap the Rn with high and well-known efficiency.

Initial measurements of a trap with a brass wool filling indicated that its trapping efficiency was less than unity at flow rates greater than about 1 liter/minute. At the low flow rate of 1 liter/minute, 10 hours would be required to pass 600 liters of gas through such a trap, an inordinately long time. The work described here was thus initiated to develop a trap filled with a sorbent with high Rn affinity that could be efficiently used at much higher flow rates.

2 CHARACTERISTICS OF THE TRAP

The trap is a U-tube made from 3/8 inch OD stainless steel tubing with 0.049 inch wall thickness. To hold the sorbent in place during high gas flow, constrictions were made near each end of the steel tube and glass wool was placed in these constrictions. This trap was installed on Rn board number 3 in the usual position of trap A.

The sorbent was supplied by John Boger. It is Chromosorb 102, mesh size 100/120, manufactured by Johns-Manville. Chromosorb 102 is a styrene-divinyl benzene polymer with very high effective surface area (300-400 m^2/g) that is mainly used for the separation of gases by chromatography. It is recommended that it be prepared for use by heating for 2 hours at

200 C under gas flow. The upper temperature limit is 250 C. The diameter of the filled trap was 7 mm and the length was about 15 cm.

This sorbent is the same material as used by the SAGE and GALLEX solar neutrino experiments to separate GeH_4 from air and from Rn by gas chromatography. The separation from radon is very important in these experiments as a significant fraction of the beta-decays that accompany Rn decay will generate pulses whose energy and rise time is identical to that of an L- or K-event from ^{71}Ge. Since any Rn added to the gas during counter filling will decay early during the counting period, these beta-decays that mimic ^{71}Ge decay will add a false signal. Most of the Rn that is present in the extracted gas sample will be separated by the chromatography, but a small fraction that may be generated during the elution will be added to the GeH_4. To maintain the GeH_4 purity and minimize the occurrence of these false ^{71}Ge signals, it is thus essential that the sorbent contain very little radium. Rn is measured during GeH_4 counting as overflow events. In a typical run about 1-3 Rn atoms are detected, so it is expected that this material will not generate significant Rn.

3 ELUTION OF RADON FROM THE TRAP

After some initial tests it became clear that the Chromosorb-filled trap could not be used in the same manner as the standard Rn-board trap A that contains brass wool. The major problem was that, if nitrogen gas had been flowed onto the trap while it was at 77 K, then, even after many minutes of pumping with the trap at 77 K, a significant volume of nitrogen gas remained on the trap. As a result, upon warming the trap to room temperature, the pressure was so great that the transfer efficiency from trap A to the liquid nitrogen-cooled trap B was poor. It was apparent that this excess nitrogen gas must be removed by pumping on the trap when it is at a temperature higher than 77 K. Some tests were then made by pumping on the trap when it was held at about -98 C, the temperature at which methanol freezes. These tests indicated that most of the nitrogen gas was released at this temperature, and that the Rn was not appreciably lost. All further experiments were then made using this procedure. (The desired temperature is obtained by pouring liquid nitrogen into methanol that is contained in a plastic beaker until considerable methanol ice has formed. To keep the trap at this temperature as it receives heat from its surroundings, additional liquid nitrogen must be frequently poured into the methanol bath.)

Although I had no direct evidence, there was also a concern that Rn would be slowly removed from the Chromosorb-filled trap at room temperature. To be certain that Rn was released by the Chromosorb, the transfer on the Rn board from trap A to the liquid nitrogen-cooled trap B was made with trap A at 100 C, obtained by placing a boiling water bath on trap A. (The water was in a glass beaker and heated by a hot plate.)

4 MEASUREMENT OF RN BOARD EFFICIENCY

Tests were then made of the efficiency of transfer on the Rn board from trap A to the Lucas cell. First a Rn sample was taken from the underground air, transferred to a Lucas cell, and counted. This Lucas cell with a known number of Rn atoms was then connected to the

Rn board at one of the usual gas inputs and the Rn sample cryopumped to the Chromosorb-filled trap A. The elution from trap A to trap B was carried out as indicated above for 30 minutes, and the transfer from trap B to the Lucas cell was made in the standard manner for 15 minutes. The same Lucas cell was used for the counting of the transferred sample as for the initial air sample. The Rn board efficiency was then calculated as the ratio of the number of atoms in the cell with the transferred sample to the number of atoms in the initial air sample, with appropriate correction for sample decay. (Counting of one of the Lucas cells after sample injection indicated that essentially all of the Rn left the cell and was cryopumped to trap A. There is thus no need to correct for residual Rn that was left over in the cell or in the tubing.)

Two such tests were made and the overall Rn board efficiency to Lucas cell 16 measured to be 69 $\pm$ 2 % and 73 $\pm$ 4 %, where the errors are only from counting statistics and background subtraction. (One departure from standard conditions occurred in the first of these measurements: during the sample transfer from trap A to trap B, trap A was at 30 C, not at 100 C as recommended above.)

The overall Rn board efficiency can be considered as the product of three factors: the efficiency of trapping on trap A multiplied by the efficiency of transfer from trap A to trap B multiplied by the efficiency of transfer from trap B to the Lucas cell. Under the conditions of these measurements it is believed that the first two of these factors are close to unity. The third factor should be approximately equal to the ratio of the active volume of the Lucas cell to the combined volume of the cell, trap B, the interconnecting parts, and the pressure transducer. Since the pressure transducer reads gauge pressure, this ratio of volumes can be determined by the following set of measurements:

1. fill trap B with ambient air at 1 atmosphere and read the pressure p_1 as indicated by the transducer,
2. evacuate trap B and again read the pressure p_2,
3. fill a Lucas cell with ambient air at 1 atmosphere,
4. close the valve before trap B and the valve after the Lucas cell port,
5. attach the air-filled Lucas cell to the Lucas cell port and again read the pressure p_5.

The transfer efficiency from trap B to the Lucas cell is then given by $1 - \frac{p_5 - p_1}{p_2 - p_1}$. Such a measurement using Lucas cells 10 and 16 (the two cells used in the measurements reported below) on the left port of Rn board 3 gave values of 68.6 and 68.3%, respectively, with an estimated systematic error of about $\pm$ 2%. The components of this error arise from the volume of air that is added when the Lucas cell is connected to the port (estimated by the quick-connect manufacturer to be as large as 0.3 cc, about 2% of the 15 cc Lucas cell volume), the volume between the Lucas cell port and the active volume of the cell, and the non-linearity of the pressure gauge.

This efficiency is in substantial agreement with the transfer efficiency measured above using Rn. It is apparent that the only substantial loss on the Rn board is in the transfer from trap B to the Lucas cell, and the overall efficiency of the board is 68 $\pm$ 2%.

5 MEASUREMENTS OF TRAPPING EFFICIENCY FROM A NITROGEN STREAM

The apparatus shown in Figure 1 was used for these measurements. It consisted of a source of flowing nitrogen gas which led either to a line which contained a Lucas cell port or to a bypass line. The gas flow then continued to the Rn board trap A and exited to atmosphere through a flowmeter.

The measurement procedure was similar to that used in determining the Rn board efficiency: Rn was extracted from the air, put into a Lucas cell, and the number of Rn atoms counted. The volume between valves B and C was then evacuated, these two valves were closed, the cell was attached to the Lucas cell port, and the Rn was injected into the enclosed volume between these valves (this volume was about 50 cc for initial measurements and 750 cc for later measurements). The Lucas cell was then removed and recounted to determine the number of residual atoms present. As this counting proceeded, nitrogen gas flow was initiated with the bypass valve A open. At some time during the flow valve A was closed and valves B and C were opened to flow the known number of Rn atoms onto trap A. At the end of the flow period, the Rn was eluted from trap A, transferred to the Lucas cell in the manner described above, and the cell was counted.

The nitrogen supply for initial measurements was from a gas cylinder; boil-off gas from a liquid nitrogen dewar was used for later measurements. In both cases the Rn content of the flowing gas was insignificant compared to the initial number of Rn atoms in the Lucas cell. So as to simulate conditions with the cover gas, the inlet pressure was at 18-20 PSI (about 1 underground atm). The trap offered considerable flow resistance so the maximum flow attainable under these conditions was 10 liters/minute.

The limited time available only permitted four such measurements to be made. Their conditions and results are given in Table 1.

Run Number	Flow Rate (l/min)	Flow Time (min)	Time when Radon Added to Gas Stream	Overall Efficiency (%)
1	8	15	5 min after start of flow	63 ± 5
5	10	15	Start of flow	67 ± 2
6	3	68	Start of flow	73 ± 5
9	10	120	15 min before end of flow	68 ± 1

Table 1: Measurements of Efficiency Under Flow.
The error given for the efficiency is the quadratic combination of the uncertainties from counting statistics and from background subtraction. For the first run the elution was made under non-standard conditions so an additional systematic uncertainty of 5% has been included.

The average efficiency for the four measurements is $68 \pm 3\%$. The overall efficiency of the Rn board is thus the same under these flow conditions as when the Rn sample was directly

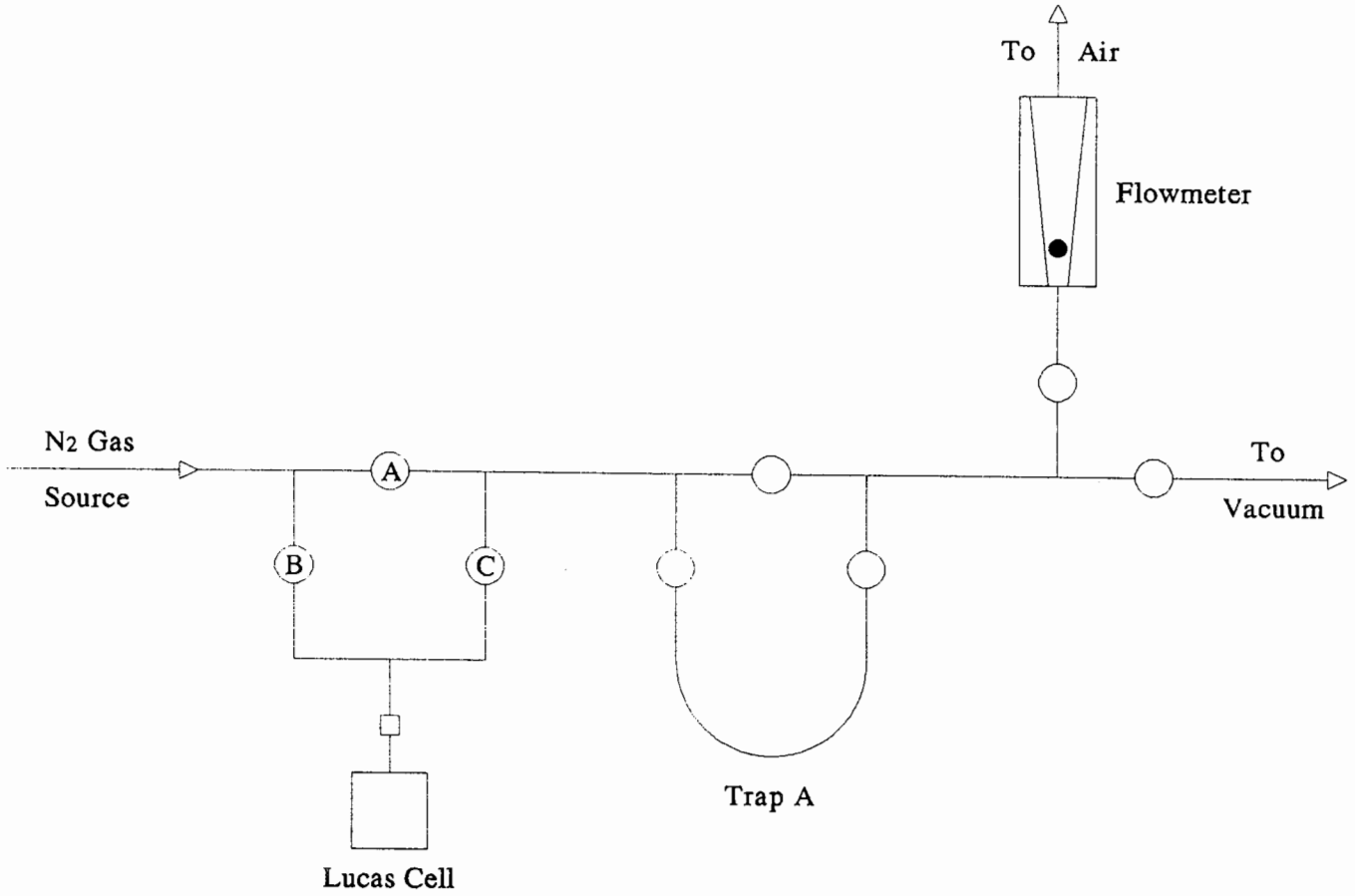

Figure 1: Schematic of apparatus used for measurement of efficiency under flow

cryopumped onto trap A. As before, the only significant sample loss was in the transfer from trap B to the Lucas cell.

Further, the efficiency was the same no matter when the Rn was added to the gas stream. This establishes that the capability of the trap to stop Rn is not reduced even after large quantities of nitrogen gas have flowed through the trap.

6 CONCLUSIONS

The trap filled with Chromosorb 102 stops Rn with approximately 100% efficiency from a stream of nitrogen at a flow rate of up to 10 liters/minute. It also retains its capability to absorb Rn even after 1000 liters of nitrogen have flowed. If the background of this trap proves to be low, as expected, then it has the capability to measure the Rn content of the cover gas at the level of 0.2 Rn atoms per liter, equivalent to a concentration in the heavy water of 10^{-15} g U/g D_2O.