The Measurement of Rn^{220} Extraction Efficiency for Various Pressures, Flow Rates, and MnO_2 Depths

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1   Introduction

Several methods have been developed to monitor the radiopurity of the light and heavy water for the Sudbury Neutrino Observatory (SNO). Common to most of these detector systems is the reliance on the widely used technique of trapping heavy elements (Radium and Thorium in particular for SNO) on MnO_2 beads. The activity is subsequently assayed by detecting the radiation resulting from the normal decay of the Radium or Thorium daughters. The problem lies in the efficient extraction of Radon gas from the MnO_2 (held in columns) prior to detection. The present report, therefore, presents the result of experiments designed to reveal the effect of pressure, bead depth and type, as well as flow rate on the extraction efficiency of the Radon emanated from spiked MnO_2. This report is also the culmination of work started by

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Karnas[1] and others before him[2][3].

2 Theory

The rational behind the design of the system presented below was rather simple. The number of atoms of the radioactive species giving rise to the detected radiation (γ particles in this case) can be found to be

$$N = N_{eq} + N_0 e^{-t\lambda}$$

where N_{eq} is the number of atoms present in the source once equilibrium has been reached after an infinite length of time has elapsed, N_0 is the initial number of atoms available to begin with, and λ, as usual, is the decay constant. The counts observed would be recorded per suitable unit of time (one hour or less) over a period of 24 hours in order to have at least 20 data points from which a curve would be plotted against elapsed time and fitted (in PAW) with a function of the form $P_0 + e^{a+bt}$. The parameter a would then be expected to be on the order of the half life of Pb^{212} - the last long lived daughter after Rn^{220}, b would be the decay constant for the data, and P_0 the equilibrium point. Thus, under different conditions, such as pressure etc., the resultant equilibrium would change accordingly. The extraction efficiency would then be a function of the equilibrium at STP and the observed equilibrium for the various conditions other than STP. Such would be the case because any pumping on the column, to create a given vacuum or pressure differential, or any flow through the column would invariably extract some of the Radon gas that is emanated from the spiked MnO_2 (Radon gas is very mobile.) The detector would subsequently register less counts when some of the activity has been removed. The extraction efficiency, η, would

[1]Scott Karnas, Centre for Research in Particle Physics, Ottawa Ontario, SNO internal publication, 1993-08-27.

[2]A.P. Ferraris, *The Extraction of Rn^{220} From Small MnO_2 Samples*, 1990-12-14.

[3]Gaëtan J.H. Laberge, *Rn Extraction by Wet and Dry Argon Gas Flow Over Th Spiked Manganese Dioxide*, Ottawa, Ontario, SNO internal publication, 1991-07-10.

simply be measured by taking the ratio of the counts observed to the equilib-
rium (at STP, and both numbers corrected for background) subtracted from
one. The last operation being due to the fact that the ratio is for the residual
activity left behind in the column and not the extracted activity. Thus, η is
defined as follows.

$$\eta = 1 - \frac{N_{eq} - BKGR}{N_{eq}(STP) - BKGR}$$

This method of determining η is insensitive to the geometry or the efficiency
of the NaI detector in the setup, provided that the column position relative
to the detector remains unchanged during a given data run.

3 System Design

In order to be able to vary pressure, flow rate, and bead depth, the apparatus
was designed as *follows*[1]. To begin with, the Argon (and subsequently
the Nitrogen) gas was supplied by a regular bottle fitted with a regulator
and a Syrfil 50mm 0.2μm pore size FN filter (to avoid any airborne solid
contaminants from accumulating in the MnO_2.) Two parallel flowmeters, of
different but complementary ranges, provided the flow rate measurements.
The flowmeters were fitted with needle valves in order to turn any one of them
off when not needed. Since the flowmeters were only calibrated at STP, a
Magnehelical meter was placed upstream such that any pressure differentials
could be eliminated by throttling the regulator found on the gas bottle. An
in-line needle valve was employed to regulate the flow instead of the valves
on the flowmeters - they only served as on-off switches. The tubing after
the needle valve was copper instead of the flexible Tygon tubing in order
to maintain a good vacuum (of about 30 mTorr) when called for. Also, to
be found on the gas input side, were two pressure gauges of complementary
ranges: an electronic gauge for 0-2000 mTorr, and a mechanical one for the
5-760 Torr range.

The activity was provided by a small amount of MnO_2 spiked with 1kBq of
Th^{228}, all of which was contained in a 1 litre acrylic column (24.5cm H by

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7.2cm D.) An Incatel vacuum pump, connected to the gas output side of the column, provided the vacuum or the pull necessary for the various operating pressures desired. The column itself was housed inside a Lead castle of 5cm minimum thickness along with a five inch diameter NaI detector (operating at a bias voltage of +1700V for one setup and -1000V for the second.) The associated electronics consisted of an amplifier and discriminator (to reduce the background contribution and avoid double counting.) The only essential differences between the two systems (two setups to carry out the experiment twice as fast) resided in the DAQ. The original setup, dubbed System1, employed a Phillips 6669 Universal Counter and PC computer for DAQ, whereas System2's DAQ consisted of a scaler attached directly to a dedicated printer.

The one litre columns were filled to any desired MnO_2 depth according to the following relation:

$$depth(cm) = \frac{dry\ Volume\ of\ MnO_2\ beads\ (cm^3)}{40.72\ cm^2}$$

where the cross-sectional area of a column is 40.72 cm^2, and the dry volume of MnO_2 is the mass of the MnO_2(dry) times a factor of 1.306. The flow rate in the column depended on the volume of the MnO_2 present and on the desired operating pressure as follows:

$$BVM(vac) = BVM(atm) \times \frac{V(vac)}{V(atm)}$$

where BVM is the number of Bed Volumes per Minute, and the volume ratio is equal to the inverse of the product of the pressure, in Torr, with a constant equal to 1.3158×10^{-3} atm/Torr (determined experimentally by Karnas.) 0.008% ?

Out of all the possible pressures, certain *values*[1] were chosen in an attempt to characterize the available range of 30mTorr to atmosphere. In the same report, authored by Karnas, a early study with various flow rates showed that the optimum was about at 100 BVM(vac) which was kept fixed from then on.

4   Data Analysis

Almost inevitably, there were some differences in the procedure born out of the changeover from Karnas to the present author. On the other hand, the effect on the data from these procedural changes seemed to be minimal. The fact that the beads were not remixed after an addition of fresh non-active beads (from the 2cm depth on) led to slightly higher extraction efficiencies for the unmixed case when at larger bead depths. This difference is most evident in the 100mm data in all four figures as the lowest data point which is for the mixed bead trial carried out in the end as a check. Also, some of the vacuum data was taken without the acrylic plug inserted inside the column's dead space above the MnO_2 bed. Although the purpose of the plug was to preclude any double counting from daughters decaying in the empty volume above the MnO_2, the effect on the data seemed negligible according to Figure 1a.

The normal sequence of measurements, after the one time straight background (with an empty column) had been done, was to determine the equilibrium level at STP, vacuum, 2Torr, 15Torr, and 300Torr. As noted in the figure captions, it was not always possible to obtain the desired pressure or flow setting. For those troublesome settings, the flow rate had to be sacrificed in order to obtain the correct pressure. As the MnO_2 volume increased, it was discovered that the vacuum pump could not operate without breaking down at 300Torr when the bead depth was 100mm. The required flow rate under those conditions was more than the pump could handle, hence the absence of all 300Torr 100mm data as is evident in Figure 2b.

Instability problems also plagued System2 at the time the 100mm bead depth was being done. In order to rectify the situation, it was necessary to change the HV power supply and operate the NaI detector at -1000V, and to reset the discriminator level as well. The 2Torr and 15Torr 100mm data in Figures 1b and 2a attest to the new found stability of System2. As might pessimistically be expected, System1 exhibited instability problems shortly thereafter and was subsequently decomissioned. Prior to this, System1 had developed a 2Torr leak on the gas input side which explains, in part, the absence of System1 data past 20mm in all figures.

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As previously mentioned, PAW was employed in the analysis of the raw data providing plots for general trends and the necessary fits for extraction efficiency determinations. In order to use the same PAW routine for analysis of cases where the general trend increased rather than decreased with time, the data was inverted by subtracting from an arbitrary constant (larger than the maximum in the data set.) The choice in constant had no more than a 0.14effect on the obtained equilibrium.

Measurements for MnO_2 on a non-porous solid bead substrate, of PS-DVB, are in progress. The available volume of the non-porous beads was only about 4cc, hence an analysis of η versus depth will not be conducted. All the beads in the 4cc sample - a mixture of three different spiking attempts of about 1.5cc each (some losses occurred) - are spiked with Th^{228} yielding a combined activity of roughly 1kBq.

Upon inspection of Figures 1a to 2b, it becomes apparent that despite the scatter of the data, there is a general trend which favors shallow MnO_2 depths. The System1 data suggests, if the System2 data is ignored for the time being, that at low depths the extraction efficiency is essentially insensitive to the flow rate. Generally, the η obtained is between 70 to 80% under those conditions. System2 data, in the same region on all figures, is consistently lower suggesting the existence of a systematic error between the two setups.

5   Conclusions

The results obtained thus far from the two systems are an improvement over the results presented in Ref. 2 and 3 which reported maximum extraction efficiencies of between 60 and 50% respectively. Overshadowing somewhat the results of the present report are the system instabilities which were not necessarily diagnosed at their onset. The subsequent variability in the data can most probably be attributed, for the most part, on the time variations in gain on the NaI detector as mentioned earlier on. Despite the preceeding considerations, it remains that the favoured conditions under which to maximize the efficient extraction of Rn^{220} are thin MnO_2 beds with flow rates

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determined only by the requirements for the individual detector systems.

optimum 10 - 100 Torr.

field thickness 1-2 cm

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220-Rn Extraction Efficiency under Vacuum

(a)

goes down

same voltag min

220-Rn Extraction Efficiency at 2Torr
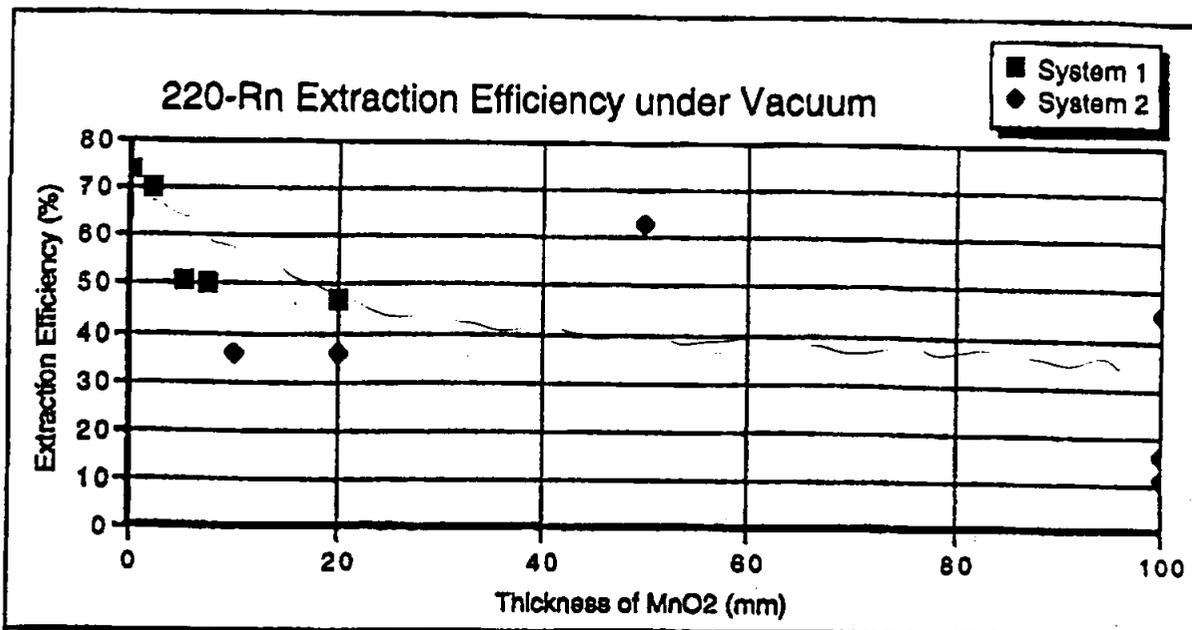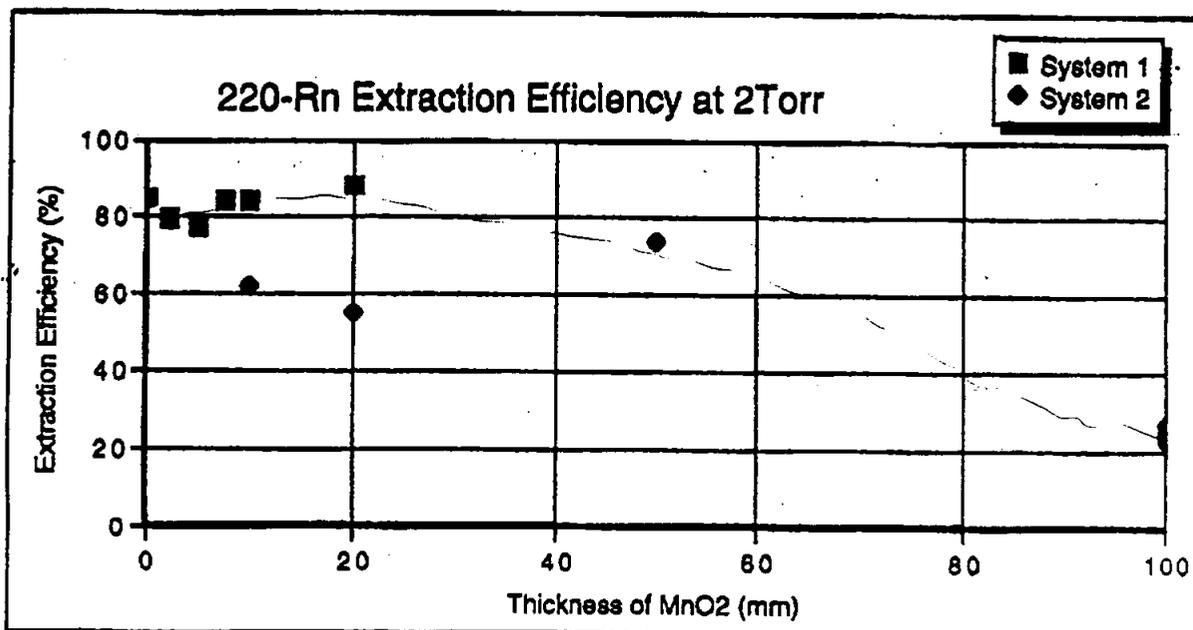
(b)

goes down

Fig. 1   Part of the data presented above is taken from Ref.1. Although most measure-
            ments were carried out with a flow rate of 100BVM(vac), it was not possible
            in some instances to do so. The 7.5mm 2 Torr measurement was actually done
            at a pressure of 5.3 Torr.   Figure 1a shows all data for vacuum measurements
            and figure 1b shows all the data for the 2 Torr measurements.

220-Rn Extraction Efficiency at 15Torr

(a)

220-Rn Extraction Efficiency at 300Torr
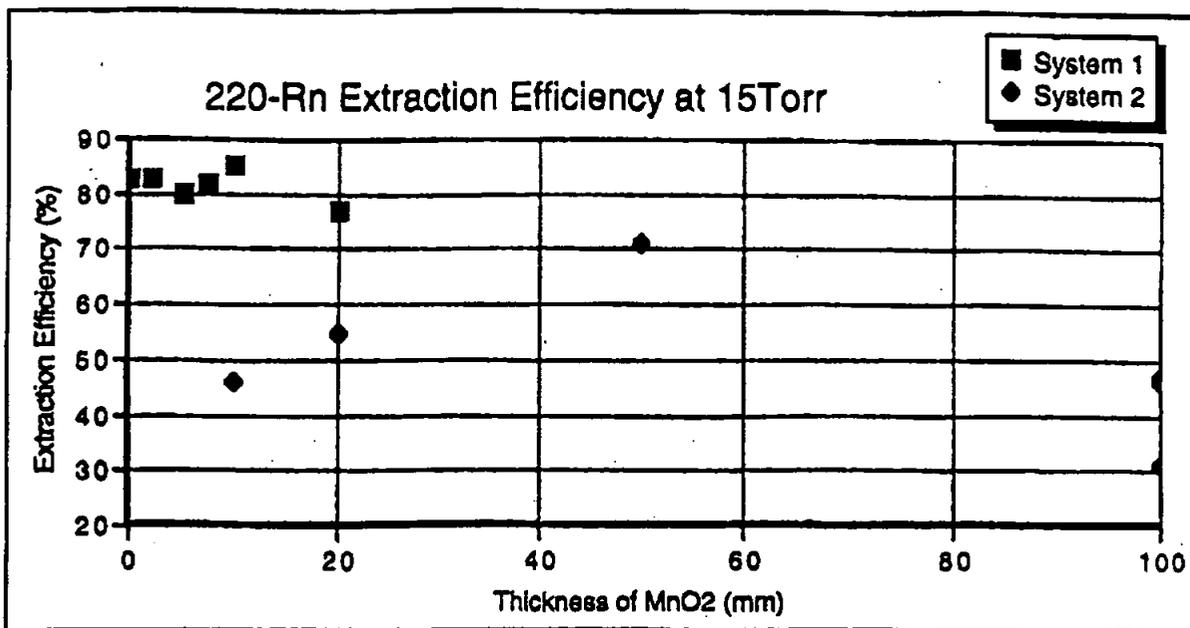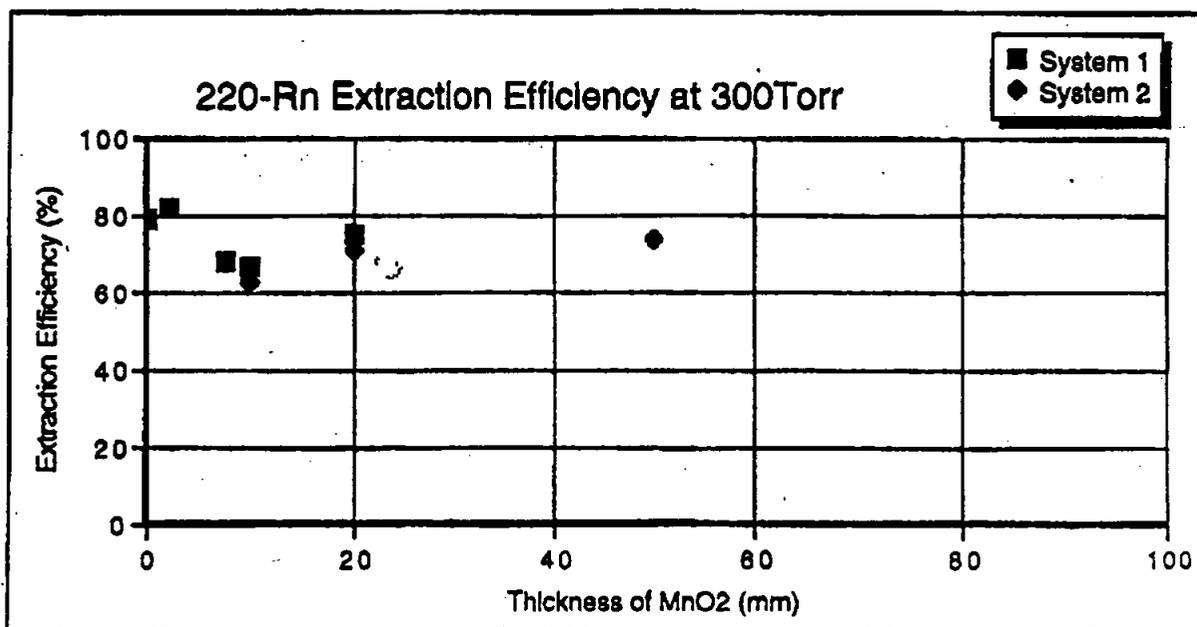
(b)

Fig. 2   Part of the data presented above is taken from Ref.1. Although most measure-
ments were carried out with a flow rate of 100BVM(vac), it was not possible
in some instances to do so. Figure 2a shows all data for the 15 Torr measure-
ments and figure 2b shows all the data for the 300 Torr measurements.