

Some Elementary Considerations about Cover Gas

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1 Activities

Some numbers to set the scale on things:

For ^{222}Rn :

1 pCi per liter of cover gas

$$= 0.037 \text{ Bq/liter} = 17527.24 \text{ atoms/liter}$$

$$= 6.519 \times 10^{-19} \text{ atoms } ^{222}\text{Rn per atom of gas}$$

For ^{220}Rn :

1 pCi per liter of cover gas

$$= 0.037 \text{ Bq per liter} = 2.07 \text{ atoms per liter}$$

$$= 7.7 \times 10^{-23} \text{ atoms } ^{222}\text{Rn per atom of gas}$$

For water:

10^{-14} g/g eq. U(or Th)

$$= 1.25 \times 10^{-7} \text{ Bq per liter (of liquid)}$$

$$= 3.38 \times 10^{-6} \text{ pCi per liter}$$

$$= 59 \text{ } ^{222}\text{Rn atoms per tonne}$$

$$= (3.3 \text{ } ^{220}\text{Rn atoms per kTonne!})$$

What is the radiopurity of the cover gas in equilibrium with H_2O or D_2O at 10^{-14} g/g eq of U/Th ?

Applying Henry's Law, this number works out to:

$$A_{\text{Rn}}^{\text{gas}} \left(\frac{\text{pCi}}{1} \right) = 7.37 \times 10^{-6} \left(\frac{k_{\text{Rn}}}{10^4 \text{ atm}} \right) \left(\frac{298 \text{ K}}{T} \right) \left(\frac{A_{\text{Rn}}^{\text{water}}}{10^{-6} \text{ pCi/l}} \right) \quad (1)$$

$$= 1.5 \times 10^{-5} \quad \text{at } T = 298 \text{ K; } k=0.6; A = 3.38$$

$$= 1.4 \times 10^{-5} \quad \text{at } T = 283 \text{ K; } k=0.4; A = 3.38$$

where A's are activities and k_{Rn} is the (temperature dependent) Henry's Law constant for Radon.

This means that if the cover gas is any dirtier than $\approx 2 \times 10^{-5}$ pCi/liter, it will contaminate the water (H_2O or D_2O) to a level worse than 10^{-14} g/g eq. U/Th.

Should or Can this level of radiopurity be achieved and/or monitored?

(a) Before the experiment can start, there is a wait period of at least a month ≈ 8 ^{222}Rn half-lives. So the initial gas can come in a factor of 256 dirtier than the above level or $\approx 4 \times 10^{-3}$ pCi/liter.

(b) Suppose the initial gas comes in typical steel cylinders (≈ 1.3 m long $\times 0.2$ m ϕ) of volume ≈ 40 liters at 2200 psi or 150 atm. These have an internal surface area of some 0.85 m^2 , and hold about 5.62 Am^3 of gas at S.T.P. If this gas has an equilibrium ^{222}Rn content due to ^{222}Rn emanation from the cylinder wall at a rate $\theta_{\text{Rn}}^{\text{cyl}}$ (probably around the generic rate of $1 \text{ m}^{-2} \text{ hr}^{-1}$), then the initial activity of this gas is:

$$A_{\text{Rn}}^{\text{gas}} @ \text{S.T.P.} = \frac{n_{\text{Rn}} \lambda_{\text{Rn}}}{V_{\text{gas}} @ \text{S.T.P.}} = \frac{A_{\text{cyl}} \times \theta_{\text{Rn}}^{\text{cyl}}}{V_{\text{gas}} @ \text{S.T.P.}} \quad (2)$$

where A_{cyl} is the internal area of the cylinder. When all is said and done, this translates into:

$$A_{\text{Rn}}^{\text{gas}} \left(\frac{\text{pCi}}{\text{liter}} \right) = 1.336 \times 10^{-6} \left(\frac{150 \text{ atm}}{P_{\text{cyl}}} \right) \left(\frac{40 \text{ liter}}{V_{\text{cyl}}} \right) \left(\frac{A_{\text{cyl}}}{1 \text{ m}^2} \right) \left(\frac{\theta_{\text{Rn}}^{\text{cyl}}}{1 \text{ m}^{-2} \text{ hr}^{-1}} \right)$$

which is more than $10\times$ better than required in the first place if the units above are of the correct magnitude. Note that ≈ 100 such cylinders will be required to provide the gas volume needed for the H_2O cover in the SNO detector alone.

(c) It is probably very hard to monitor a ^{222}Rn level of 10^{-5} pCi/liter in a gas and harder still to measure this level of ^{220}Rn or a factor of 10 less ^{222}Rn . 10^{-5} pCi/liter of gas means a ^{222}Rn concentration of 0.18 atoms/liter and a ^{220}Rn concentration of 2.07×10^{-5} atoms/liter. To measure this, one would have to flow 100 liters of gas through a Radon trap, then transfer to a Lucas cell and count the 18 atoms of ^{222}Rn at a count rate of $\approx 3 \times 3$ cnts/day. Counting the ^{220}Rn or a level 10 times less ^{222}Rn is virtually impossible.

first day

22 gas pbt
sup chain
that too

100% eff

2 D₂O Cover

Surface Area of D₂O in detector = 1.77 m²

Surface Area of 2 m of acrylic chimney = 9.5 m²

Surface Area of (3 m)³ glove box = 54 m²

Volume of cover gas = 30 m³

Can one maintain the required purity of the cover gas in this (small) volume ?

If the total surface areas A in the cover gas volume emanate ²²²Rn at a rate θ_{Rn} , and if the cover gas is static, then the number of ²²²Rn atoms which will accumulate in it is:

$$n_{Rn} = 8000 \left(\frac{A}{60 \text{ m}^2} \right) \left(\frac{\theta_{Rn}}{1 \text{ m}^{-2} \text{ hr}^{-1}} \right) \quad (3)$$

as opposed to the number which is in equilibrium (from Henry's Law) due to U in the water:

$$n_{Rn} = 8100 \left(\frac{V_{gas}}{30 \text{ m}^3} \right) \left(\frac{k_{Rn}}{0.6 \times 10^4 \text{ atm}} \right) \left(\frac{298 \text{ K}}{T} \right) \left(\frac{A_{Rn}^{D_2O}}{10^{-14} \text{ g/g eq. U}} \right) \quad (4)$$

These numbers are probably pessimistic because θ_{Rn} for acrylic from which presumably the glove box is constructed is at least a factor of 10 lower than what is taken above. In any case, radon emanation rates from sources etc. which occupy the glove box should be checked.

Besides, the total number of ²²²Rn atoms in the D₂O is about 60000. So even if all the extra 8000 atoms from the glove box were to go into the D₂O, it would increase the total ²²²Rn in it by only 13%.

Can one quantify the equilibrium conditions for the radon gas in the cover gas—water system ?

Yes! If the radon concentrations in the cover gas and water (or D₂O) are in equilibrium, they are distributed according to Henry's Law. If there are n_x^g and n_x^l atoms of gas species x in the gas and liquid respectively and the total amount of liquid and gas are given by the number of moles n_l and the gas volume V respectively, then all these quantities are related via the ideal gas law and Henry's Law according to:

$$P_x = \frac{n_x^g RT}{V} = k_x X_x = k_x \frac{n_x^l}{n_l} \quad (5)$$

where P_x is the partial pressure of x in the gas volume, and X_x is (approximately) the mole fraction of x dissolved in the liquid. If one adds an amount dn_x^{tot} of atoms to this system, they redistribute themselves in the gas and liquid:

$$dn_x^{tot} = dn_x^g + dn_x^l \quad (6)$$

The relation between dn_x^g and dn_x^l is found by differentiating (5) so that:

$$dn_x^g \frac{RT}{V} = dn_x^l \frac{k_x}{n_l} \quad (7)$$

Combining (6) and (7), and doing algebra, one finds how the additional dn_x^{tot} atoms redistribute themselves in the liquid and gas phase:

$$dn_x^l = dn_x^{tot} \left(\frac{1}{1 + \frac{k_x V}{RT n_l}} \right) = dn_x^{tot} \left(\frac{1}{1 + y} \right) \quad (8)$$

and the complimentary quantity

$$dn_x^g = dn_x^{tot} \left(\frac{y}{1 + y} \right) \quad (9)$$

with of-course:

$$y = \frac{k_x V}{RT n_l} \quad (10)$$

For Rn in D_2O , $k_{Rn} = 0.6 \times 10^4$ atm, at $T = 298$ K, with $n_l = 1$ kT, this works out to $y = 0.1325$, so that 88% of all the Rn atoms go into the D_2O and 12% into the cover gas as promised before *if equilibrium is achieved*. This still points to the necessity of keeping a fairly tight rein on the radio-purity of the cover gas.

What about air leaks, D_2O losses, contamination due to airlocks etc. ?

I don't know at what rate outside air is likely to diffuse into the cover region. If the cover region is kept at slightly above ambient room air pressure, the radio-contamination should be minimized. Vapor Loss depends on the achievable leak rate. Should be worked out. Even if the leak rate is as large as 1 torr-liter s^{-1} , if the D_2O temperature is $10^\circ C$ so that the vapor pressure in the cover region is ≈ 10 torr, a simple calculation shows that

we lose about 415 g of D_2O a year. Efforts should be made to minimize this leak rate.

Consider a typical airlock of volume 1 m^3 . If the D_2O sits at 10°C , its vapor pressure is ≈ 10 torr, while the mine air pressure is at ≈ 1.25 std. atm or 1000 torr. So each time the airlock is opened, we lose 1 Am^3 of cover gas and 0.01 Am^3 or ≈ 10 g of D_2O . Over the life of the experiment, if there are 1000 openings, we'll lose a total of 10 kg D_2O , a truly insignificant amount. But each time this is done, there must be a supply of clean cover gas to purge the airlock, or else the D_2O will get contaminated by radon.

The single biggest vapor loss/ vapor recovery effort will occur when the acrylic tank is filled or emptied. If this is a static process with respect to the cover gas, ie. it is simply displaced, at 25°C , there is 1000 m^3 of gas with a partial vapor pressure of ≈ 25 torr or 0.033 atm. This contains a total of 26 kg D_2O which needs to be recovered, but only twice—once during fill, and once during the final empty sequence. The actual required recovery effort will depend on the details of the fill and empty sequence.

3 H_2O Cover

Surface area of H_2O in SNO detector $\approx 340 \text{ m}^2$

Volume of H_2O cover gas $\approx 500 \text{ m}^3$ Other surfaces exposed to cover gas are:

SS liner $\approx 100 \text{ m}^2$

Top (plastic + SS ??) $\approx 340 \text{ m}^2$

Some numbers:

Using the numerology developed already, we find that the H_2O at a level of 10^{-14} g/g eq. U, contributes a total of 1.5×10^5 atoms of ^{222}Rn to the 500 m^3 of cover gas, while at this level, 7300 tonnes of it itself contains about 4.3×10^5 atoms of ^{222}Rn . The H_2O is likely to be dirtier than this, so both numbers may go up by a factor of 3-10. From this one may deduce that any addition of Rn to the gas will eventually redistribute itself in a 70:30 ratio between water:gas. If the total of $\approx 500 \text{ m}^2$ of surfaces in contact with the cover gas emanate ^{222}Rn at the rate of $1 \text{ m}^{-2} \text{ hr}^{-1}$, then this will add in equilibrium, a total of 6.6×10^4 atoms of ^{222}Rn , of which 70% will

end up in the water, an increase of 15% over the 10^{-14} g/g eq. U level.

More exactly:

If we solve (10), for $V = 500 \text{ m}^3$, $n_l = 6 \text{ kT}$, we find $y = 0.37$ which means that if equilibrium is reached, 73% of all Rn atoms go into the water, and 27% stay in the cover gas.

Is equilibrium likely to be reached? While the D_2O is more or less static, we are told that the H_2O is moving at (relatively) high velocities, so that the "turnover" time is about 10 hours! Does this mean that equilibrium in the gas—water solution can be reached in a time scale ≈ 10 hours? If so, it raises the interesting possibility of removing ^{222}Rn from the water via the cover gas! If *all* the ^{222}Rn can be removed from the water via its interaction with the cover gas in a characteristic time, $\tau_{\text{circulation}} = 10 \text{ hr}$, then the reduction in ^{222}Rn concentration would be $\tau_c/\tau_{\text{Rn}} = 0.076 \approx 0.1$ ie. 90% of the ambient ^{222}Rn could be removed. Note that this is the *only* way of reducing the ambient Rn levels in the H_2O since its recirculation time (≈ 35 days) through the purification loop is too long compared to the ^{222}Rn decay time (5.5 days).

Is Rn removal via cover gas feasible?

Maybe! There are several practical impediments. The details of how much Rn actually gets removed depends on how closely equilibrium is achieved, ie. on the details of water flow patterns, condition of the water surface etc. as well as on the radiopurity or flow rate of the cover gas (which carries off the ambient Rn).

(1) The conditions for equilibration can be enhanced by agitating the water surface. An extreme case would be to install sprayers just below the surface, so the surface water is broken up into small droplets. The total water flow rate through these sprayers would need to be very high—since we are talking about a recirculation time of 10 hrs for 6 kT of water, to take advantage of this, we need to spray at the rate of 10000 liters/min; however the pressure head for the sprayer pumps would need to be very small (3 or 4 feet of water). There may be easier and better ways of agitation.

(2) The cover gas would have to be recirculated and purged of Rn. From equations derived elsewhere, we find that to achieve 90% degassing at a water flow rate of 10000 l/min, we need a gas throughput rate of $10 \text{ Am}^3/\text{min}$ at a gas pressure of 1 atm. Can the gas be cleaned by passing at this rate through a room temperature Granulated Activated Charcoal [GAC]

column? If the GAC has to be at low temperature (-60°C) then the recirculating cover gas will first have to be dehydrated or it will clog up the cold GAC. Since the GAC has to be cooled anyway, it would make sense to use a heat exchanger and cold trap at the same temperature to dehydrate. If the initial cover gas contains 10 torr of water vapor (at 10°C), then the total cooling power required to condense the H_2O and cool the (assumed monoatomic) cover gas at a flow rate of $10 \text{ Am}^3\text{s}^{-1}$ is 7 kW at low temperature (-60°C ?). On the other hand if room temperature GAC is efficient at removing Rn, it would suffice to simply put some *in situ* in the cover gas region without the need for recirculating the cover gas. This GAC would then have a long time scale (10 hrs) available to remove the ^{222}Rn in the cover gas.

In summary

- (1) The possibility of removing ambient ^{222}Rn via cover gas exists. Theoretically, as much as 90% removal can be achieved. Even if only a factor of 2-4 removal is achievable, this may be valuable.
- (2) The amount of ^{222}Rn removal via cover gas depends critically on the convective recirculation time of the H_2O .
- (3) The removal can be facilitated by agitation of the water surface. This agitation has to be at a high rate.
- (4) The practicality of the method is limited by the requirement for keeping the cover gas which is the carrier, free of ^{222}Rn . This requirement can be trivially accomplished by room temperature *in situ* GAC. However the ^{222}Rn removal efficiency and cleanliness of room temperature GAC needs to be established.