

Some Aspects of the Radon Problem  
=====

E.Bonvin, B.Cleveland, P.Jagam,  
H.Lee, V.Novikov, M.Steiner

June 6, 1996

1. INTRODUCTION	...	1
2. On BACKGROUND of CLOSED LOOP	...	3
3. On BAGGING OPTION	...	6
4. On RADON LOAD from URYLON	...	8
5. CONCLUSIONS	...	9
6. References	...	9
7. APPENDIX 1	...	10
8. APPENDIX 2	...	11

## 1. INTRODUCTION.

=====

Throughout this report we'll often refer to a concept of Rn diffusion, so, an introduction is to be of some use.

For a system of two volumes separated by a membrane, equilibrium (steady-state) Rn flux through a unit area of the membrane is written as [1]

$$F = \frac{2 D S}{DL} \times \frac{(C_1 - C_2)}{[\exp(d/DL) - \exp(-d/DL)]}, \quad (1.1)$$

where

- $C_1, C_2$  - external Rn concentrations on each side of the membrane,
- $D$  - diffusion constant,
- $d$  - membrane's thickness,
- $DL = \sqrt{D \tau}$  - diffusion length, where  $\tau = 4.8 \times 10^5$  s is Rn lifetime,
- $S$  - solubility of the membrane; it is define as the ratio of the surface concentration in the membrane to external concentration.

The radon diffusion and solubility constants can be measured by means of so called "time-lag" method (see [2] and references therein for details).

To avoid corrections of the diffusion and solubility coefficients regarding Rn decay in the membrane, the requirement of the diffusion time through membrane to be much smaller than Rn lifetime should be met (i.e. thickness of the membrane  $d$  should be much smaller than diffusion length  $DL$ :  $d \ll DL$ ).

Fig.1 [2] illustrates the behaviour of the total amount diffused as a function of time:

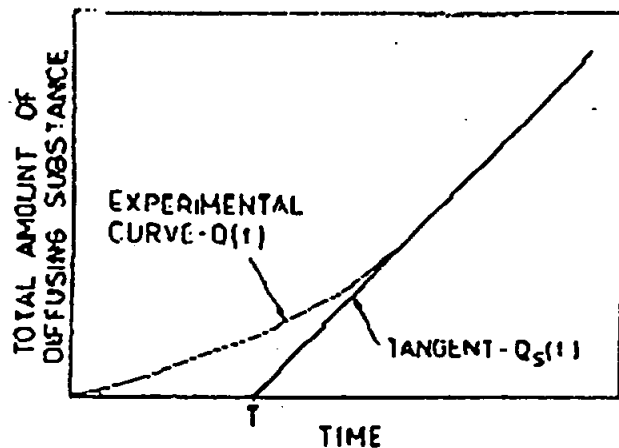


Fig. 1. Approach to steady-state flow through the membrane.

The slope of the straight line is the steady-state flow rate,

$$F = \frac{D \times S \times C_1}{d} \quad (1.2)$$

As one can see, (1.2) is just an extrapolation of (1.1) at  $d \ll DL$  and  $C_2 \ll C_1$  ( $C_2$  is usually maintained low compared to the opposite face). The intercept of the straight line on the  $t$ -axis is referred to as the "time-lag":

$$T = \frac{d^2}{6D} \quad (1.3)$$

$T$  defines a time required for a steady-state flow of  $R_n$  through a membrane to be reached.

To measure  $D$  and  $S$  separately [2,3] is somewhat more difficult than to measure just its product,  $K = D \times S$ , called permeability.

Diffusion and solubility constants (measured for a thin membrane) can be used for calculations of  $R_n$  reduction by a thicker membrane made of the same material; evaluation of  $R_n$  reduction properties from the knowledge of permeability only is limited for a membrane of the same thickness which was used in the measurements of permeability.

2. On BACKGROUND of CLOSED LOOP.

=====

If water in system MDG+closed loop (see [4] for definition) is continuously degassing, the steady-state flow of Rn into the system will be reached sooner or later. Moreover, the actual measured Rn flux can tell us whether it is due to diffusion only (if properties of membranes involved are known, such as O-rings, pumps' diaphragms, ...) or some leaks are also involved, or, otherwise, roughly estimate membranes' properties (if the system is known to be leak tight, - as was done in [5]).

Water in the MDG+closed loop was degassed over a period of about 8 days, with 5 extractions done. The purpose of the experiment is to verify if the amount of Rn extracted from the closed loop is consistent with the possibility of Rn diffusion through pumps' membranes, as a source of Rn in closed loop.

Table 2.1 Experimental details.

	Action starts @ abs time	Action's duration, h	Details of the extractions	Dura- tion, h	LC #	Counts /time, s	Counts/day, corrected for eff.)
Degas/Extr_1	0.0	7.3	5	8	469/82446	472 +/-27	
Stopped		19.3					
Degas/Extr_2	26.6	5.5	4	9	222/70689	380 +/-31	
Stopped		38.0					
Degas/Extr_3	70.0	10.5	5	7	347/117892	299 +/-19	
Stopped		14.5					
Degassing	95.0	10.3					
Stopped		12.5					
Degas/Extr_4	118.0	11.3	5	8	220/70618	269 +/-20	
Stopped		18.1					
Degassing	146.5	6.9					
Stopped		14.8					
Degas/Extr_5	168.0	6.2	5	7	185/69675	269 +/-22	
End	174.2						

By continuous degassing the water, Rn concentration in the closed loop (C<sub>2</sub>) has been significantly reduced. This, however, should not have much affect on the initial Rn flux into the closed loop because membranes and O-rings were continuously exposed to air (C<sub>1</sub>). In addition, C<sub>2</sub> << C<sub>1</sub> prior to the experiment. Therefore, from (1.1) one should expect that the steady-state Rn flow into the closed loop should have been reached in a quite short time after the beginning of the experiment, and, in fact, it should rise with time.

Thus, we've concluded that the observed drop in the Rn rate is likely due to slow removal of the "original" Rn from the system.

Out of 269 counts/day (net) in a Lucas cell after a 5 h extraction, 115-120 are due to MDG alone. Therefore, approx 150 counts/day is a contribution of the closed loop. This rate corresponds to a total Rn flow inside closed loop of

$$F_{\text{meas}} = \frac{150}{0.167 \times 0.62 \times 3 \times 0.75 \times 5} = 138 \text{ h}^{-1}, \quad (2.1)$$

For the calculations, the required numbers used were: duration of extraction run (5 h), Rn transport efficiency in the MDG (0.75), number of alpha-particles emitted per one Rn atom in a Lucas cell (3), detection efficiency of the Lucas cell (0.62), fraction of Rn atoms decayed in the first day of counting (0.167).

Potential sources of Rn inside closed loop are diffusion through O-rings [5] and diffusion through pumps' membranes driven by compressed air which is "full" of Rn (see also Appendix 1 for pumps' details):

Table 2.2 Pumps' specifications.

- # of pumps in closed loop	2
- # of membranes per pump	2
- membrane's thickness:	0.46 cm
including:	
- teflon	0.13 cm
- polyurethane	0.33 cm
- membrane's surface	200 cm <sup>2</sup> (at most)
- compressed air pressure	approx 3 atm
- air consumption per pump	8 m <sup>3</sup> per hour
- Rn content in pump's air	3 pCi/l (or 180 Rn atoms/cm <sup>3</sup> )

To estimate Rn load from membranes, one needs to know properties of teflon and polyurethane; here are data available:

Table 2.3 Polyurethane (PU) and teflon Rn diffusion properties.

	Permeability, x10 <sup>7</sup> , cm <sup>2</sup> s <sup>-1</sup>	Diffusion constant, x10 <sup>7</sup> , cm <sup>2</sup> s <sup>-1</sup>	Solubility
teflon [3]	3.5	-	-
PU [2]	2.3	0.41	5.6

As one can see, properties of teflon and PU are somewhat similar; for this reason (as well as because of lack of necessary data on D and S for teflon) we'll deal PU membrane with combined thickness of PU+teflon (0.46 cm).

From (1.1) one can find Rn flux per unit membrane's surface (d=0.46 cm, C<sub>1</sub> = 180 cm<sup>-3</sup>, C<sub>2</sub> = 0.):

$$F_{\text{calc}} = 2.2 \times 10^{-5} \text{ cm}^{-2} \text{ s}^{-1}, \quad (2.2)$$

which means Rn load of 63 per hour per pump.

Table 2.4 summarizes estimated Rn loads into closed loop, compared with the measured number. We do not know if there is a significant contribution from Rn diffusion through O-rings of valves located in dead legs, as well as from a membrane used to smooth water pressure (similar to a pump's

membrane), so, "at least" and "at most" estimations are given (for a simplified diagram of the closed loop. see Appendix 2).

Table 2.4 Rn load in closed loop.

Rn source	Radon load, Rn/h
from pumps	126
from valves - at least (2x1" + 2x2" valves in the circulation line)	13
- at most (1/2" + 6x1" + 2.5x2" in the circulation line and dead legs)	27
from a membrane (used to smooth water pressure)	0
- at least	32
- at most	
TOTAL Calculated:	
- at least	139
- at most	185
MEASURED (see 2.1):	138

Numbers in the table are approximate; expected effect from the pumps might be smaller if teflon is less permeable to Rn than polyurethane (there is a concern that permeability numbers in [3] are systematically high).

As one can see, most of Rn in closed loop is expected to come from the pumps. To verify this statement, a similar experiment was proposed, with pumps driven by Rn-free compressed N<sub>2</sub> instead of air; it is, however, seems unreasonable because of large amount of N<sub>2</sub> required (estimated air consumption is 6-8 m<sup>3</sup>/h per pump, [6]).

Fortunately, the fact that it takes quite long time for Rn to diffuse through membranes ( "time-lag" T = 10 days, estimated from (1.3) ) gives us a chance to find out if radon from pumps (if any) is due to diffusion. The proposed steps are as follows:

- air supply to the pumps is well flushed by N<sub>2</sub>, and is filled by N<sub>2</sub> at 60 psi or so;
- for 10 days or so the system (MDG+closed loop) is not touched; air supply is flushed by N<sub>2</sub>, from time to time (not very critical if no flushing at all is done);
- water in the MDG+closed loop is degassed for at least 10 h, and couple of extraction is done, 5 h long each. Before running the MDG, it is advisable to measure Rn content in N<sub>2</sub> in the air supply line.

If the measured rate is below 180-200 day<sup>-1</sup>, it would support the possibility of high Rn flux through membranes.

### 3. On BAGGING OPTION.

=====

To reduce Rn load into water system from valves, bagging option was proposed. The idea is to create, inside the bag, lower Rn concentration than the one outside of the bag.

Rn concentration inside the bag depends on properties of the bag's material as a Rn barrier, as well as on geometry (bag's surface and volume which it covers).

What to expect?

-----  
 There are some useful data available [3,7] on Rn permeability.

Table 3.1 Rn permeability of different materials.

	Permeability, cm <sup>2</sup> s <sup>-1</sup>	
	[3]	[7]
Mylar	1.6x10 <sup>-8</sup>	-
Aluminized mylar	5.2x10 <sup>-8</sup>	2.8x10 <sup>-9</sup>
Al foil	4.6x10 <sup>-8</sup>	-
Polyester	5.3x10 <sup>-8</sup>	-
Polyethylene (high density)	1.1x10 <sup>-8</sup>	-

Equilibrium Rn concentration inside the volume (V) covered by bag of surface Surf is written as

$$C_2 = \frac{F \times Surf}{V}, \quad (3.1)$$

where F is the Rn flux per unit surface of the bag (1.1). C<sub>2</sub> is driven by Rn concentration C<sub>1</sub> outside the bag (approx 3 pCi/l, or 60 Rn atoms per cm<sup>3</sup>). In the test experiment with O-ring stack, Surf=5000 cm<sup>2</sup>, V=1.5 l, thickness d =0.02 cm. Rn diffusion properties of the used material have not been measured, so, we'll use numbers from Table 3.1. (As only permeability is given, we'll give estimations for 2 arbitrary values of solubility: S=1. and =5.).

Table 3.2 Rn reduction inside the mylar, aluminized mylar or polyester bag.

Thickness	Permeability taken from: (see table 3.1)	Solubility	Time-lag, h	Rn reduction (i.e. C <sub>1</sub> /C <sub>2</sub> )
0.02 cm	[3] Al mylar or polyester	1.	0.4	1.0
		5.	1.8	1.0
	[7] Al mylar	1.	6.6	2.8
		5.	33.1	3.3
	[3], mylar	1.	1.2	1.0
		5.	5.8	1.0
	[3] polyethylene	1.	1.7	1.0
		5.	8.4	1.0

Rn reduction numbers are rather low and strongly depend on performance. (Note, however, the concern about validity of data on permeability given in [3]). Therefore, the bagging option can be successful only if:

- proper and thick enough material is chosen for the bag (diffusion constant D and solubility S should be known to be low!);
- bagging geometry has a good (large) volume/surface ratio.

Measurements.

As mentioned above, the O-ring stack was bagged (approx 5,000 cm<sup>2</sup> of surface; 1.5 l volume of air inside); bag is a 3 layer material: polyethylene, polyester and aluminum, of total thickness of 0.02 cm), and the extractions were made. Results of the extractions are presented in Table 3.3, together with results of extractions from the un-bagged stack.

Table 3.3 Extractions from the O-ring stack.

##	Seal time	Extraction time	LC#	Counts /Time	Counts/day, corr. for eff.
Without bag:					
1. (Nov_29)	21 h	40 min	8	283/65111	366 +/-25 *)
2. (Dec_7)	8 days	25 min	8	521/83361	530 +/-29 **)
3. (Dec_15)	8 days	60 min	2	443/66778	664 +/-40
4. (Dec_20)	20 h	55 min	8	167/75845	180 +/-16
5. (Jan_5)	16 days	60 min	8	372/86440	362 +/-22 ***)
With bag:					
6. (May_21)	5 days	31 min	9	127/53854	225 +/-24
7. (May_30)	9 days	40 min	9	260/98981	252 +/-20
8. (June_3)	4 days	41 min	2	113/55124	193 +/-21
9. (June_4)	20 hours	40 min	8	73/91669	59 +/- 8

- \*) As the stack was set up in the previous day (Nov\_28), there is a significant contribution in Rn rate from outgassing.
- \*\*\*) Still there may be some contribution from outgassing.
- \*\*\*) Prior to the extraction the stack was placed inside a bag, and N2 was continuously flushed inside the bag for 24 h.

We see Rn reduction factor of about 3 for the O-ring stack with bag, compare to the un-bagged stack. As there is an uncertainty in the strength of Rn source (mine air), so, one has to perform special measurements in order to give more accurate number.

Calculations predict no Rn reduction in the O-ring stack for the bag made out of material we're using (this may be due to some systematics in [3] mentioned earlier); the factor 3 is close to what one would expect from aluminized mylar [7]. If the material we're using is close in properties to aluminized mylar ( K = 2.8x10<sup>-9</sup> cm<sup>2</sup> s<sup>-1</sup>), we can predict Rn reduction of 6-12 if DOUBLE bagging is used:

Table 3.4 Rn reduction expected from double bagging.

Thickness	Solubility	Rn reduction
0.04 cm	1.	6.4
	5.	12.3



4. On RADON from URYLON.  
=====

Radon concentration in a room with internal Rn source (emanation, ... ) and in the presence of ventilation is written as [8]

$$C = \frac{\frac{LOAD}{V}}{L_{rn} + L_{vent}} + \frac{C_{ext}}{1 + L_{rn}/L_{vent}}, \quad (4.1)$$

where

- C - radon concentration in the room of, m<sup>-3</sup>,
- C<sub>ext</sub> - radon concentration in incoming air, m<sup>-3</sup>,
- V - room's volume, m<sup>3</sup>,
- LOAD - internal Rn load, Rn atoms per hour,
- L defines Rn removal by decay (L<sub>rn</sub>) and ventilation (L<sub>vent</sub>):
- L<sub>rn</sub> = 0.0076 h<sup>-1</sup> - Rn decay constant,
- L<sub>vent</sub> - air exchange ventilation rate (number of room volumes per hour).

In the SNO case, two similar equations can be written, for inner (1,700 t) and outer (5,300 t) water volumes (as R/O removes Ra very effectively, down to 10<sup>-14</sup> g/g of U equiv., we neglect here by Rn contribution from Ra in the water):

$$C_1 = \frac{\frac{F_{urylon} \times S}{V_1}}{L_{rn} + L_{v1}} + \frac{\frac{LOAD_{eman1}}{V_1}}{L_{rn} + L_{v1}} + \frac{C_2}{1 + L_{rn}/L_{v1}}, \quad (4.2)$$

and

$$C_2 = \frac{\frac{LOAD_{eman2}}{V_2}}{L_{rn} + L_{v2}} + \frac{C_1 \times (1 - E_{degas})}{1 + L_{rn}/L_{v2}}, \quad (4.3)$$

where

- C<sub>1</sub> - Rn concentration (m<sup>-3</sup>) in the internal water,
- C<sub>2</sub> - Rn concentration (m<sup>-3</sup>) in the external water,
- V<sub>1</sub> = 5,300 t - volume of external water,
- V<sub>2</sub> = 1,700 t - volume of internal water,
- L<sub>v1</sub> = 0.0017 h<sup>-1</sup> - "ventilation" rate for the external water (150 l/min x 60 min / 5,300 m<sup>3</sup>),
- L<sub>v2</sub> = 0.0053 h<sup>-1</sup> - "ventilation" rate for the internal water (150 l/min x 60 min / 1,700 m<sup>3</sup>),
- E<sub>degas</sub> = 0.98 - Rn degassing efficiency of the water by the Process Degasser [9],
- LOAD<sub>eman1</sub> = 76,000 h<sup>-1</sup> - Rn load rate into external water from all Rn emanation sources ([10], upper limit),
- LOAD<sub>eman2</sub> = 6,840 h<sup>-1</sup> - Rn load rate into internal water from all Rn emanation sources ([10], upper limit),

9

FLUX\_urylon x S - Rn load into water from urylon coating. Here  
FLUX\_urylon is Rn flux from urylon,  $m^{-2} h^{-1}$ ,  
and  $S = 2,000 m^2$  - surface covered by urylon.

A simple relation between  $C_2$  and  $F_{urylon}$  follows from (4.2) and (4.3)

$$C_2 = 0.55 \times 10^{-3} \times F_{urylon} + 0.54, \quad (5.4)$$

where Rn concentration  $C_2$  is represented now in units  $10^{-13}$  g/g of equilibrium U concentration.

As an example, SNO can afford Rn flux from urylon as high as 833 per  $m^2$  per hour if the required purity of the inner water is  $10^{-13}$  g/g U equiv.

## 5. CONCLUSIONS.

=====

- Rn diffusion through pumps' membranes is likely to be a major source of radon in the closed loop. An experiment is suggested, to verify this statement.
- Bagging the O-ring stack reduces Rn concentration by a factor of about 3 inside the stack. Double bagging would reduce Rn concentration by approx 10 times.
- A simple relation between Rn flux from urylon and Rn concentration in the inner water is derived. As an example, if SNO can afford Rn concentration of  $10^{-13}$  g/g U equilib., Rn flux from urylon should be  $< 833 m^{-2} h^{-1}$ .

## References.

=====

- [1] B.Sur, SNO-STR-92-056.
- [2] M.Wojcik, NIM B61 (1991) 8.
- [3] J.Bigu, E.D.Hallman and L.Kendrick, SNO-STR-91-069.
- [4] A.Hamer, H.Lee, V.Novikov, "Water in the Monitor...", Oct 19, 1995.
- [5] E.Bonvin, P.Jagam, H.Lee, V.Novikov, J.Tan, SNO-STR-96-013.
- [6] K.McFarlane, Private Communication.
- [7] A.Hallin, Private Communication.
- [8] A.Nero, Physics Today, April 1989, p.32.
- [9] A.DeKok, A.Hamer, H.Lee, A.Noble, V.Novikov, SNO-STR-96-016.
- [10] M.Liu, H.Lee, A.McDonald, NIM A329 (1993) 291.

APPENDIX

From POTATO@hincks.physics.carleton.ca Wed May 29 10:09:16 1996  
Date: Wed, 29 May 1996 10:02:14 -0400 (EDT)  
From: POTATO@hincks.physics.carleton.ca  
: novikov@hincks.physics.carleton.ca  
: POTATO@hincks.physics.carleton.ca  
Subject: Re: Update of the "Lilac Review"  
Resent-Date: Wed, 29 May 1996 10:02:16 -0400 (EDT)  
Resent-From: novikov@hincks.physics.carleton.ca  
Resent-To: novikov

Hi Vladimir,

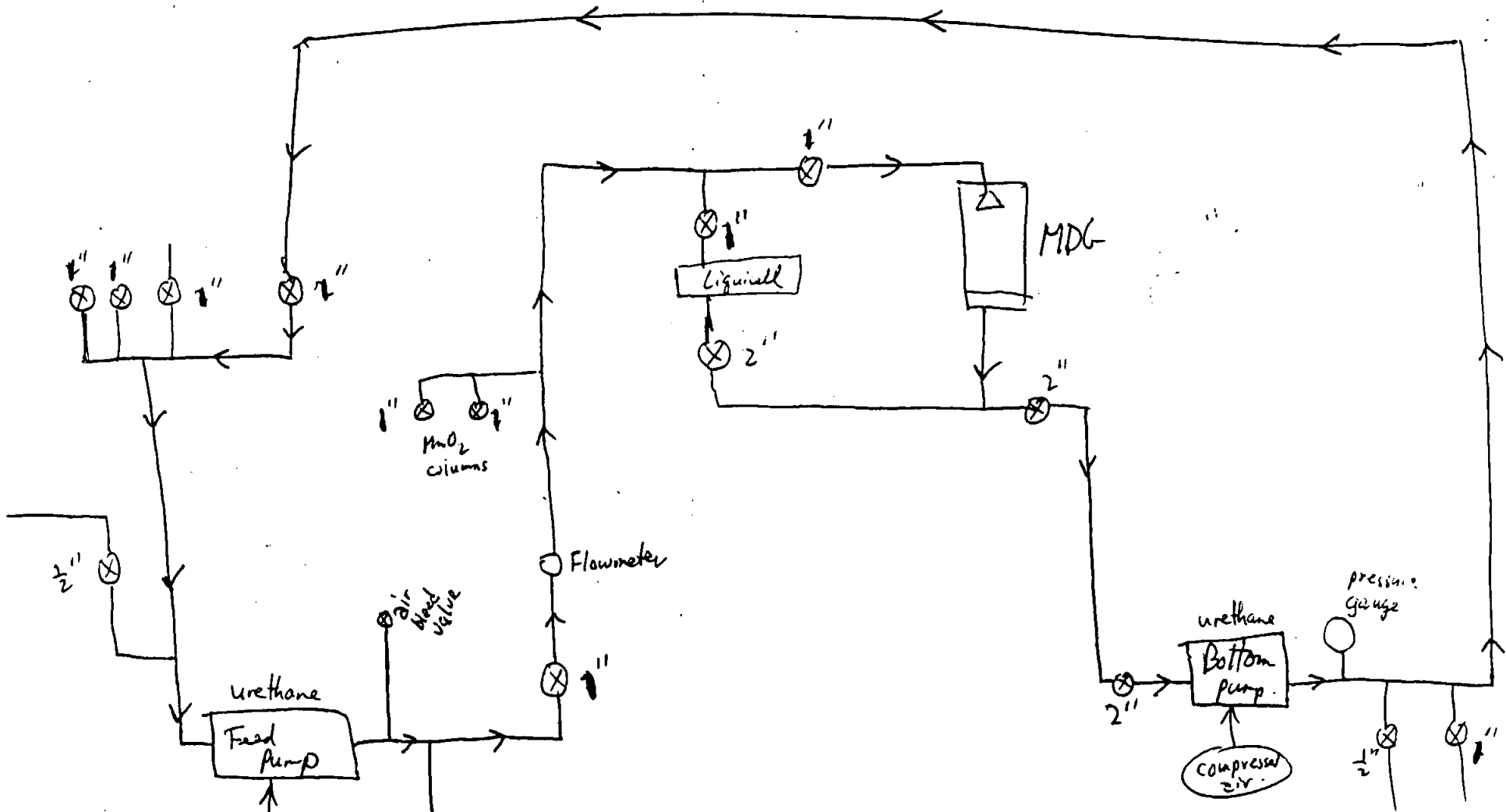
The Polyurethane has a thickness of 0.130 +- 0.001 inches.

The Teflon has a thickness of 0.051 +- 0.001 inches

The exposed area I have to guess a bit.

There is a flat flange where the seal is made. This has an outside radius of about 3.93 +- 0.02 inches inside radius of about 3.26 +- 0.02 inches so I guess the area exposed uses this radius. There is a hole at the center of each diaphragm where I suppose there is a washer or similar. In the polyurethane, the center hole has a diameter of 0.80 inches, whereas the teflon's is 0.49 inches.

I hope this helps, Cheers, Tony



Urethane  
Big diaphragm here  
to smooth water  
pressure

⊗ = Valve, Most valves  
are 1".

APPENDIX 2