Physics Considerations of Rope Radioactivity - Implications for Rope Procurement -

Jan M. Wouters
Los Alamos National Laboratory
Los Alamos, NM 87545

March, 1994

Introduction:

This report briefly summarizes the original SNO requirements for rope radioactivity and explores the cleanliness implications resulting from a cleaner acrylic vessel and heavy water. The status of the radioactivity measurements on Kevlar and Vectran are presented. The report concludes with a discussion of possible options with respect to the filament, my choices, and a plan of action for obtaining and testing rope.

SNO Requirements:

How much thorium and uranium contamination is tolerable in the rope is dictated by two factors: the size of the background signal that can originate from the rope and how much rope is near the acrylic vessel. This section examines the permissible background signal from four different points of view. First, what are the requirements according to the white book and subsequent evaluations [see SNO-STR-90-153, SNO-STR-91-019, SNO-STR-91-025, and SNO-STR-92-**(P. Skensved, B. Robertson, W. Frati "Rope Background Implications")]. Second, what are the requirements based on the current measured radioactive contamination of the acrylic vessel and heavy water. Third, how do the white book requirements change if ^{3}He detectors are used to measure the neutral current flux. Fourth, how do they change if a cleaner vessel and heavy water are used with the ^{3}He counters. In the following discussions, I will assume that the background signal originating from the rope should be small, that is less than 20%, compared to that from the acrylic vessel.

White Book: The white book (page 75) sets maximum levels for thorium and uranium in the acrylic (i.e. 1.9 ppt and 3.6 ppt, respectively) based on what was thought to be achievable. Such a level of contamination results in approximately 6.5 neutrons/day (SNO-STR-92-**) produced in the heavy water of which 45% are captured producing light when using NaCl (Mike Lowry – MCNP calculation). Thus the background introduced by the acrylic vessel is comparable to that originating from contaminants in the heavy water (~3 neutrons/day for 0.011 ppt Th, most of which do not escape). This "acrylic induced" background will occur within one meter of the vessel (~95% see white book page 94). Using these approximate values Frati and Beier have analyzed how well the various solar signals can be extracted. Their key message is that a SSM neutral current rate of ~14 events/day (~5000 per Kt-yr) can be measured to 5-10% accuracy.

Using the white book values we can calculate the maximum allowable thorium and uranium quantities for the rope. The weight of the acrylic vessel is 71,900 lbs (32,640 Kgs) which results in a maximum permissible amount of thorium and uranium of 62 μg and 118 μg, respectively. Since publication of the white book several key factors have changed that require adjustment of these two values. First, the acrylic background neutron capture rate in D_2O + NaCl was 24% in the white book (page 95) and is now 45% (see above). Second a decrease of about 9% is needed because the original design specification was for a 30,000 Kgs acrylic vessel. The revised numbers are 30 μg (0.94 ppt) and 57 μg (1.79 ppt) for thorium and uranium, respectively. (One side observation is that the background signal originating from thorium is approximately 12 times larger than from the same quantity of uranium. Thus 30 μg of thorium contributes almost 6 times more to the background than 57 μg of uranium.) Using our 20% criteria we obtain 6.0 μg and 11.4 μg as the maximum amounts of thorium and uranium that the rope can contain.

Expected Acrylic Vessel: Radioactivity measurements of acrylic destined for inclusion in the acrylic vessel have averaged 0.05 ppt thorium and < 0.25 ppt uranium (SNO-STR-93-042). If we make the assumption that during construction of the acrylic vessel the average thorium concentration increases to 0.25 ppt (private communication Davis Earle) then the rope can only have 1.6 μg of thorium if we keep to our 20% criteria.

At 1.9 ppt thorium the acrylic vessel contributes approximately half the neutral current background using NaCl. At 0.25 ppt and with the heavy water expected to be less than 0.006 ppt the total background is reduced by roughly a factor of 4. However, as was pointed out earlier with a SSM signal of 14 neutrons/day, a heavy water background of 3 neutrons/day, and an acrylic vessel background of 3 neutrons/day, one can already measure the neutral current rate to ~5-10%.

Tables 1&2 present the results of a calculation, based on statistical arguments only, which determine the change in the uncertainty of the neutral current flux in going from white book thorium and uranium values to the above values for the acrylic and heavy water. The rope is assumed to have 350 ppt Th and 700 ppt U which produces 0.42 neutrons/day (Peter Skensved – private communication) and which is a level that is probably achievable. In Table 1 the calculation for salt-out yields a charged current uncertainty of 40 / 1120 = 3.6% (ignoring the neutral current contribution). For salt-in the corresponding error is of 50 / (1120 + 860) = 2.8%. Combining these results gives an error of 7.9% for the neutral current flux. In Table 2, the improved case, the results for salt-out and salt-in are: 38 / 1120 = 3.4% and 47 / (1120 + 860) = 3.4%, respectively. The derived neutral current signal has an error of 7.2%. The improvement is approximately 10%.

Table 1: Error in signals using white book values.

	White book	Assumptions[4]	Salt-out 1/2 yr		Salt-in 1/2 yr	
	(1/2 Kt-yr)		Det. Signals	%Err	Det. Signals	%Err
Total N	-		1496	2.6	2479	2.0
C.C.	2800	1/3 SSM	1120	3.6[1]	1120	2.8[2]
N.C.	2500	SSM	270		860	
D_2O bkgd	605	0.011 ppt Th + U	65	12.4	208	6.9
NaCl bkgd	180	1.3 ppt Th + U	-	-	62	12.7
Acrylic bkgd	1190	1.9 ppt Th + 3.6 ppt U	38	16.2[3]	214	6.6[3]
Rope Bkgd	78	350 ppt Th + 700 ppt U	2.5		14	

Table 2: Error in signals using factor 2 improvement in D_20 and factor of 8 improvement in acrylic contamination levels.

	White book	Assumptions[4]	Salt out 1/2 yr		Salt in 1/2 yr	
	(1/2 Kt-yr)		Det. Signals	%Err	Det. Signals	%Err
Total N	-		1430	2.6	2189	2.1
C.C.	2800	1/3 SSM	1120	3.4[1]	1120	2.5[2]
N.C.	2500	SSM	270		860	
D_2O bkgd	303	0.0055 ppt Th + U	33	17.5	104	9.8
NaCl bkgd	180	1.3 ppt Th + U	-	-	62	12.7
Acrylic bkgd	158	0.25 ppt Th + 0.5 ppt U	5.1	36.0[3]	28	15.3[3]
Rope Bkgd	78	350 ppt Th + 700 ppt U	2.6		14	

[1] N.C. signal is ignored in salt out option. Error is obtained from propagation of errors assuming background and total N signal are statistics limited.

[2] C.C. and N.C. signals use summed error obtained from propagation of errors assuming that background and total N signal are statistics limited.

[3] Acrylic and rope backgrounds used summed error.

[4] Assumptions: With salt out neutron capture efficiency is 27% for neutron originating from neutral current signal, or D_2O background, and 8% for neutron originating from acrylic or rope. With salt in the appropriate values are 86% and 45% respectively. Detection efficiency for a neutron once captured is 40%.

White Book with ^{3}He Counters: The situation improves with use of ^{3}He counters because the neutron capture efficiency is lower near the acrylic vessel which enhances the neutral current signal relative to the (γ,n) background originating from the acrylic vessel. Comparing the detection efficiency of (γ,n) with respect to neutral current signal we obtain a ratio of ~0.5 for NaCl and only ~0.3 for ^{3}He (MCNP - Mike Lowry, neutron transport calculation - Jerry Wilhelmy private communication). Thus, one could allow the contamination in the acrylic vessel to increase by 2/3 with a corresponding increase in the rope to 10.0 µg and 19.0 µg thorium and uranium.

Expected Acrylic Vessel with ^{3}He Counters: Again if the acrylic vessel is a factor of roughly 8 better than the white book (see section *Expected Acrylic Vessel*) then we have to reduce the rope contamination levels to 2.7 µg and 5.2 µg thorium and uranium, respectively.

We can repeat the statistics analysis done in Tables 1 and 2. The neutral current error is 68 / 2350 = 2.9% for the normal detector while it is 58 / 2350 = 2.5% for the cleaner detector – an improvement of ~14%. (See Tables 3 and 4). It should be noted that in just one year of running the ^{3}He detectors out perform the NaCl by over a factor of 2 in this simple calculation.

Table 3: Error in signals using white book values and ^{3}He counters.

	White book (1 Kt-yr)	Assumptions[3]	Det. Signals	%Err
Total N	-		3468	1.7
N.C.	5000	SSM	2350	2.9[1]
D_2O bkgd	1210	0.011 ppt Th + U	569	4.2
^{3}He det. bkgd.	360	Assume same as for NaCl	169	7.7
Acrylic bkgd	2380	1.9 ppt Th + 3.6 ppt U	357	5.1[2]
Rope Bkgd	155	350 ppt Th + 700 ppt U	23	

Table 4: Error in signals using ^{3}He counters with factor of 2 improvement in D_2O and factor of 8 improvement in acrylic contamination levels.

	White book (1 Kt-yr)	Assumptions[3]	Det. Signals	%Err
Total N	-		2874	1.9
N.C.	5000	SSM	2350	2.5[1]
D_2O bkgd	605	0.0055 ppt Th + U	284	5.9
^{3}He det. bkgd.	360	Assume same as for NaCl	169	7.7
Acrylic bkgd	315	0.25 ppt Th + 0.5 ppt U	47	11.9[2]
Rope Bkgd	155	350 ppt Th + 700 ppt U	23	

[1] N.C. signal uses error obtained from propagation of errors assuming that backgrounds and total N signal is statistics limited.

[2] Acrylic and rope backgrounds used summed error.

[3] Assumptions: Neutron capture efficiency is 47% for neutron originating from neutral current signal, D_2O background or ^{3}He detector background and 15% for neutron originating from acrylic or rope.

Implications for Rope Radioactivity: Two components contribute to the amount of rope near the acrylic vessel. The first is the length of rope in the rope groove which is 160 cm. The second is the length of rope in the two vertical members of each rope. These vertical members do not contribute uniformly because as one progresses away from the equator of the acrylic vessel the amount of shielding provided by the light water increases. Using the transmission equation for gamma rays $I/I_0 = \exp\{-x\mu\rho\}$, where x = distance, $\mu = 0.045$ cm^2/g, $\rho = 1.0$ g/cm^3, and the known geometry between any position on a vertical member and the shortest distance to the acrylic vessel from that point, one can calculate the effective length of the rope if it contributed background/unit length at the same rate as the rope in the rope groove. (This calculation omits the change in solid geometry which would decrease this effective length.) The additional length is 146 cm per leg or 452 cm total effective length per rope. For ten ropes made of Vectran with an average weight of 24.2 lbs/100 ft (11.0 Kg/3048 cm) we obtain a total weight for the rope contributing to the background of 16.3 Kg. For Kevlar the weight is about 15% more because a slightly larger diameter rope is required.

For each of the cases discussed above we can calculate the concentration levels. These values are listed in Table 1. One factor which can lead to a slight relaxation of these numbers (~12%) is that a gamma ray originating in the rope has to traverse on average twice as much acrylic as a gamma ray originating in the acrylic. Comparing to the acrylic produced background of 6.5 neutrons/day, we obtain 0.53 neutrons/day for rope with 350 ppt Th which compares well with the 0.42 neutron/day value obtained from the Queen's Monte Carlo (see above).

Table 5: Contamination limits in rope for several cases assuming rope contributes 20% as much background as acrylic vessel. (Vectran weight used for ppt columns).

Case	Th (μg)	U (μg)	Th (ppt)	U (ppt)
White book with revisions	6.0	11.4	370	700
Expected acrylic vessel	1.6	3.1	100	190
White book (rev.) with 3He counters	10.0	19.0	610	1160
Expected acrylic vessel with 3He counters	2.7	5.2	165	320

Summary: Meeting the requirements of the "expected acrylic vessel" will require rope with no more than ~100 ppt Th. As will be shown in the next section, this concentration is lower than we can routinely measure and is lower than the average measured so far for both Vectran and Kevlar filaments which presently are both at 350 ppt Th or below. **The arguments presented above indicate that even if the rope has 350 ppt Th, the expected improvement beyond the white book in the acrylic and heavy water caused backgrounds will not lead to a significant improvement in the neutral current measurement.** Thus, for the moment, the overriding reason for improving the cleanliness of the rope is not based on the physics that will be derived, but on the lower anticipated backgrounds originating from the acrylic vessel and heavy water.

Radioactivity Results

Two methods have been used to measure a majority of the thorium and uranium concentrations in filament; these are direct gamma counting (DGC) and neutron activation analysis (NAA). The direct counting facilities consist of the 3 crystal detector located at the 4600' level at Sudbury which is jointly operated by Laurentian University and The University of Guelph, and the Merlin detector located at the Oroville dam which is operated by the Lawrence Berkeley Laboratory. (Sudbury experiments started with a single crystal detector that could not achieve the required sensitivity.) Neutron activation analysis has been done by CRL, The University of Guelph, and LANL. Key results for Vectran are summarized in Table 2a while those for Kevlar are summarized in Table 2b.

Table 2a: Summary of Vectran filament radioactivity measurements.

Laboratory	Type	Th (ppt)	Comments
Sudbury	DGC	<700 ± 93%	~1 Kg sample, Single crystal detector
Sudbury	DGC	<284 ± 30%	14.1 Kg, Prelim.
LBL	DGC	<360 (1 s)	2.6 Kg
LBL	DGC	178 ± 114 (1 s)	4.0 Kg, Prelim.
U. Guelph	NAA	362 ± ~11 (530, 629, 230, 465, 130, 189)	Ave of 6 meas., error average of individual errors.
CRL	NAA	245 ± ~55 (310, 180)	Ave of 2 meas., error average of individual errors. Sample prepared by U. Guelph
LANL	NAA	213 ± 60 (1 s)	Single measurement only

Table 2b: Summary of Kevlar filament radioactivity measurements.

Laboratory	Type	Th (ppt)	Comments
Sudbury	DGC	<800 (1 s)	~1 Kg sample, Single crystal detector
Sudbury	DGC	229 ± 170 (2 s)	18.8 Kg
LBL	DGC	<300 (1 s)	
LBL	DGC	<133 ± 98 (1 s)	4.3 Kg
U. Guelph	NAA	91± ~16 (146, 40, 87)	Ave of 3 meas., error average of individual errors.
CRL	NAA	<72; <700; <1000	Samples prepared by U. Guelph
CRL	NAA	350 ± 120	Sample prepared at CRL using new tech.
LANL	NAA	144 ± 100% (1 s)	Single measurement

Direct Gamma Counting: A sensitivity of less than 200 ppt is obtainable with the present configurations of the Sudbury and LBL detectors. The Sudbury detector uses three ~50% efficient crystals and a very large sample size ~16 Kg to achieve the desired sensitivity. LBL relies on a single 100%+ efficient detector using a reasonably large sample size (i.e. ~4.0 Kg). The present results have the Vectran filament below 250 ppt Th and possibly below 150 ppt. The Kevlar filament results are less conclusive with a 350 ppt measurement from Sudbury and a <120 ppt measurement from LBL.

Direct gamma counting has two distinct attributes. First by looking at several transitions including the 2614 KeV transition in the decay of ^{212}Po one is sensitive to the end of the decay chain. In addition, by counting large samples, any variability in the filament is averaged out.

Neutron Activation Analysis: The NAA results for filament have displayed wide variability. There was a major discrepancy between CRL and the University of Guelph which I believe is now resolved. Even so results from a single group (e.g. see results from University of Guelph for Vectran, Table 2a) have shown wide variability. Numerous measurements are thus required to have any confidence in the measured concentrations. The apparent difficulty with NAA is the combination of small sample size (i.e. 1 – 10 g) and the very high surface area of the filament which facilitates contamination.

On the positive side NAA has a sensitivity that is ~50 ppt which is at least a factor of two if not three better than DGC. In addition, the turn around time and throughput for NAA is much better which will be a concern during rope production.

The best present NAA results are those from the University of Guelph because they used large samples sizes (i.e. ~10 g as opposed to the ~1 g samples of CRL and LANL), have several measurements, and have results that are reasonably reproducible. These measurements on both Vectran and Kevlar agree reasonably well with those from DGC.

Summary: For the time being I have higher confidence in the DGC than the NAA analysis. Even so we can only state that Vectran is probably less than 250 ppt Th while Kevlar is 350 ppt or less. There is good evidence that the Kevlar is lower than 350 ppt when the DGC measurement from LBL is combined with the University of Guelph and LANL NAA measurements. The implications for this level of contamination is that the filaments will probably meet the “revised white book” and “revised white book + ^{3}He counter cases”, but will not meet the “Expected acrylic vessel” case. In the latter case the rope contribution to the neutral current background will approximately equal that due to the acrylic vessel. A more complete report on the filament radioactivity is in preparation.

Discussion:

This section briefly summarizes the arguments for choosing Vectran or for choosing Kevlar.

Vectran: Vectran is the only filament that we could clean up because Hoechst-Celanese is willing to use our suggestions to modify their production facility. Indeed, Hoechst-Celanese has recently started making a medical grade Vectran which is essentially the same material, but with an extensive quality control program for cleanliness.

Very early NAA measurements indicated that Vectran polymer just prior to spinning is quite clean. The most believable measurements are 84 ± 4 ppt Th from Guelph and a recent 70 ± 5 ppt Th from CRL. There are a number of other measurements from CRL, but these occurred prior to resolving the NAA problem. CRL has also done a single mass spectrometer measurement yielding 35 ppt Th, however, there were significant difficulties in dissolving the polymer for this measurement.

The obvious place to clean up the filament is during the spinning stage when the most surface area is exposed to the environment. We could install a hepa filtered system that blows air across where the filament comes out of the spinneret and is gathered into a yarn.

With respect to engineering properties, Vectran has essentially zero creep at a given load. However, this lack of creep in the filament does not imply a corresponding lack of creep in the rope which can elongate through mechanical relaxation. Vectran also has superior braid resistance, which is probably not a concern for the acrylic vessel support ropes.

Kevlar: Kevlar, apart from the one DGC measurement from Sudbury, appears to be quite clean; potentially cleaner than Vectran. On the negative side approximately 15% more Kevlar is required than Vectran because Kevlar snags and breaks easily during twisting and braiding. Vectran is quite supple. Kevlar also has roughly 10 times less K than Vectran which has K levels of 60-80 ppm. On the other hand, Kevlar has significantly higher levels of other contaminants many of which appear related to the elements found in steel.

From an engineering stance Kevlar has a long history and so its properties over a long period of time are well known. Though it does creep, this creep is approximately 0.1% at 10% load after a single year which is small.

A clear advantage of Kevlar is its cost which is ~1/5 that of Vectran. Thus the bids for comparable test and production ropes for SNO differ by a factor of 2.2.

Choices:

Selection of Manufacturer: Beginning with four manufacturers we have received two final bids from Yale Cordage and Samson Ocean Engineering. The choice is clear, based on price and ability to deal with an unusual order, I would select Yale Cordage.

Choice of rope construction: Two rope designs exist. One is a 2 ply, 12 braid, high braid angle rope with a roughly 1.3" diameter, while the other is a 1 ply 12 braid, low braid angle rope with a roughly 0.9" diameter. The latter rope is clearly superior for our use because it is engineered specifically to support a large dead weight using the minimal amount of material. Its creep properties also promise to be vastly superior which is important in keeping the acrylic vessel accurately positioned.

Material choice: Kevlar. Kevlar is cheaper, it can do the job mechanically, its cleanliness is probably very similar to Vectran, and we do not need a cleaner rope to do the physics.

Plan of Action:

Obtaining a rope sample. We must obtain a rope sample soon!!! Such a rope sample is critical to uncovering any problems with respect to radioactivity that the rope making process may cause. I would obtain Kevlar rope samples from Yale Cordage using the RFQ already bid upon that includes a provision for purchase of test rope.

Test program - mechanical: The rope needs to be placed in an acrylic test jig to verify that mating between rope and acrylic vessel. This test must also include loading the rope to see how the acrylic behaves. Three short ropes need to be tensioned to destruction to verify the mechanical strength.

Test program - radioactive: A large sample of rope must be sent to Sudbury and LBL for DGC. Smaller samples must undergo NAA to obtain an independent verification of the rope cleanliness and to measure intermediate samples for determining where contamination might be introduced during the rope production process.

Polymer measurements: To determine if cleaning up the Vectran filament is even feasible in the eventuality that Kevlar proves too dirty we must measure additional samples of Vectran polymer using NAA. I propose to send 10 samples, already prepared, to CRL while measuring 6 at LANL.