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NEW $^{232}U/^{228}TH$ GAMMA SOURCE FOR TRITIATED WATER MONITOR

INTRODUCTION

The $^{232}U/^{228}Th$ source from Isotopes Products, Inc, has been received and tested for operation with the tritiated water monitor.[1-8] This source has an effective half-life of 73 years and is an attractive replacement for the reactor activated ^{24}Na sources, which have half-life of only 15 hours. The tests described in this report appraise the adequacy of this source by comparing its performance to that of ^{24}Na used in the earlier studies. The new source has already been used successfully to assist recovery operations following the tritium release on September 2, 1984.[9-10] It is now apparent that the monitor is ready to be turned over to SRP for routine use.

SUMMARY

The new $^{232}U/^{228}Th$ source is very suitable for the tritiated water monitor. The background from (α,n) reactions in the source is too small to have a serious effect on detection limits, and somewhat longer counting times can be used to rectify this if extremely low-level detection limits are ever required. The source should have adequate activity for well over 100 years.

The monitor itself will be turned over to SRP in the near future. A schedule for this turnover has been developed.

DESCRIPTION OF TESTS

The tests were performed similarly to those discussed earlier.[2,8] These tests calculated a detection limit for D_2O using the formula

$$\Delta N_D = \Delta R/GS \qquad (1)$$

where

ΔN_D = D_2O detection limit (μL)
ΔR = $D_2O(\gamma,n)$ minimal detection count rate (cpm)
G = Gamma source strength (mCi)
S = Probe detection sensitivity (cpm/μL/mCi)

The value of ΔR is calculated using the formula based on nuclear counting statistics (see appendix), viz

$$\Delta R = 2\sqrt{R_b/t} + 1/t \qquad (2)$$

where

R_b = Background count rate (cpm)
t = Count time (min)

With a ^{24}Na source R_b is simply the room background R_r. For the $^{232}U/^{228}Th$ source a contribution from (α,n) needs to be included; thus,

$$R_b = R_r + AG \qquad (3)$$

where

A = Alpha-produced neutrons per source (cpm/mCi)

Here we note that equation (3) is general if we use A = 0 for the Na-24 sources.

By substituting equations (2) and (3) into equation (1), we obtain a detection limit which is consistent with both that of the earlier work with ^{24}Na and the present studies with $^{232}U/^{228}Th$. The resulting formula is:

$$\Delta N_D = \frac{2\sqrt{(R_r + AG)/t} + 1/t}{GS} \qquad (4)$$

This formula is used for all tests discussed in this report.

TEST COMPARING ^{24}NA AND ^{232}U/^{228}TH SOURCES

The small probe geometry $A_{\parallel}$ [2,8] was used to compare the two types of sources. In this geometry, the Na glass or the U/Th rod (0.125 in. diam x 2.0 in. long) is centered on the largest side of the probe and parallel to the neutron detectors. Then a polyethylene capsule with 10 ml D_2O is placed next to the source. Respective background counts R_b and R_r were taken with the D_2O sample removed and then with the source removed. The background corrected countrate for the D_2O sample R_D was used to calculate the sensitivity as $S = R_D/(10\ ml)/(source\ mCi)$. Furthermore a value of A was calculated as $A = (R_b - R_r)/(source\ mCi)$. The resulting parameters S and A were then used in equation (4) to calculate the probe detection limits.

The detection limits for typical source magnitudes and counting times are compared in Table I, for the ^{24}Na and ^{232}U/^{228}Th sources. In the past, ^{24}Na sources ranged from 1 to 7 mCi, while the ^{232}U/^{228}Th source was delivered as a nominal 3 mCi of ^{228}Th, which will buildup to 9 mCi before decaying with a half-life of 73 year. Considering these ranges and the fact that counting time can be increased somewhat, the results in the Table show that the U/Th source can perform just as well as the Na sources have in the past. Also, more recent measurements[10] demonstrate that judicious positioning of the source can decrease the (α,n)-background substantially, allowing shorter counting times to be used.

TESTS FOR DIFFERENT PROBE GEOMETRIES WITH U/TH SOURCE

Using the same procedure as in the preceding section, detection limits for various probe geometries were examined. The small probe was used in geometry $A_{\perp}$, and an even smaller new probe was examined in addition. This new probe also uses smaller detectors and can be placed about 1/2 in.- and 1 in.-diameter stainless steel pipes, which are typical in 234-H. The aim of the new probe is to permit measurements in more confined spaces. Unfortunately, as the probe is made smaller the detection efficiency also decreases.

Results for the different probes are summarized in Table II, for the ^{232}U/^{228}Th source. It is noted that the new smaller probe does not have detection limits as low as those of the old probe. They are about 5 times higher. Some of the disadvantage can be made up by counting longer. This lesser performance is not due to the source, but due to the smallness of the probe. The small probes were calibrated using D_2O samples placed in stainless steel pipes, which also cause a decrease in detection efficiency.

CONCLUSIONS AND PROJECTIONS

The tests indicate that the $^{232}U/^{228}Th$ source can be used instead of the ^{24}Na sources. Even though the ^{24}Na sources do offer the better background conditions (A = 0) and thus more favorable detection, this advantage is only marginal and can be rectified by longer counting. Indeed, because preparation of ^{24}Na sources typically has required a 24-hour lead time, somewhat longer counting time (which is not likely to exceed 20 minutes) will have no impact on the turnaround time for the measurements. In fact, the new $^{232}/^{228}Th$ source will provide a quicker turnaround, even when such longer counting is required. The most striking advantage of the $^{232}U/^{228}Th$ source is its ready availability. The ^{24}Na source does require a reactor to produce it, and there have been occasions where no SRP reactors were available for this service. The possibility of using outside reactor services (e.g. Ga. Tech.) have been explored; however, use of these tends to increase the lead time for producing ^{24}Na.

The 73-year effective half-life of the $^{232}U/^{232}Th$ is clearly an advantage over the 15-hour half-life of ^{24}Na. In addition to being a "permanent source", the $^{232}U/^{228}Th$ source does not require the user to keep track of decay corrections during the day(s) of the measurements, as is the case with the ^{24}Na source.

It should be appreciated that the ^{228}Th decay chain includes Tl-208 which provides the 2614 keV gamma that acts in the (γ,n) reaction on deuterium. The actual $^{232}U/^{228}Th$ source received from Isotopes Products Inc is composed nominally of 10 mCi ^{232}U and 3 mCi of ^{228}Th. Because ^{232}U decays with a 73-year half-life to ^{228}Th which has a 1.9-year half-life, the ^{228}Th will eventually be in equillibrium decay with its buildup from ^{232}U and thus have an effective half-life of 73 year. A graph of the ^{228}Th activity as a function of time is shown in Figure 1. Note that the ^{228}Th activity will grow from 3 mCi to a maximum of 9 mCi in about 10 years and then gradually decay back to 3 mCi after an additional 121 years. Essentially then, the source, which already has been appraised as acceptable at 3 mCi, should be suitable for at least another 131 years or up to the year 2115.

TURNOVER OF OPERATIONS FROM SRL TO SRP

The tritium monitor will be turned over to 234-H personnel in the near future. The turnover will include (1) the equipment and guidelines for constructing modified probes for special applications, (2) complete set of drawings, purchase orders, and equipment manuals for maintenance purposes, and (3) a set of written procedures for efficient and safe use of the mmonitor. A schedule for this turnover is given in Table III. A training session will be included to demonstrate the procedures.

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TABLE 1. Comparison of $^{232}U/^{228}Th$ and ^{24}Na Sources

Each gamma source in the comparison below is evaluated for same probe geometry ($A_{\|}$ with old probe). The detection limits are determined from equation (4), using experimental measurements of S, A, and R_r.

^{24}Na Source: S = 18.4 cpm/mCi/ml
A = 0.0 cpm/mCi
R_r = 7.0 cpm

Detection Limits (μL of D_2O)

^{24}Na mCi	Count Time (min) 10	20	30	40	50	60
1	96	67	54	47	42	38
2	48	34	27	23	21	19
3	32	22	18	16	14	13
4	24	17	14	12	10	10
5	19	13	11	9	8	8
6	16	11	9	8	7	6
7	14	10	8	7	6	5
8	12	8	7	6	5	5
9	11	7	6	5	5	4

$^{232}U/^{228}Th$ Source*: S = 28.7 cpm/mCi/ml
A = 10.5 cpm/mCi
R_r = 7.0 cpm

Detection Limits (μL of D_2O)

^{228}Th mCi	Count Time (min) 10	20	30	40	50	60
1	96	67	54	47	42	38
2	60	42	34	30	26	24
3	47	33	27	23	21	19
4	39	28	23	19	17	16
5	35	24	20	17	15	14
6	31	22	18	16	14	13
7	29	20	16	14	13	12
8	27	19	15	13	12	11
9	25	18	14	12	11	10

* S and A were deduced using the nominal 3 mCi value of ^{228}Th, per Isotope Products, Inc. Plans for obtaining an accurate measured value are being explored, for future refinement of the results.

TABLE II. $^{232}U/^{228}Th$ Source Performance*

Old Probe - Geometry $A_{\parallel}$: See previous table

Old Probe - Geometry $A_{\perp}$: S = 30.2 cpm/mCi/ml
A = 4.3 cpm/mCi
R_r = 7.0 cpm

Detection Limits (μL of D_2O)

^{228}Th mCi	Count Time (min)					
	10	20	30	40	50	60
1	74	51	42	36	32	29
2	43	30	24	21	19	17
3	32	23	18	16	14	13
4	27	19	15	13	12	11
5	23	16	13	11	10	9
6	21	14	12	10	9	8
7	19	13	11	9	8	8
8	17	12	10	9	8	7
9	16	11	9	8	7	6

New Probe - 1/2 in. SS pipe: S = 7.8 cpm/mCi/ml
A = 8.0 cpm/mCi
R_r = 7.0 cpm

Detection Limits (μL of D_2O)

^{228}Th mCi	Count Time (min)					
	0	20	30	40	50	60
1	327	228	186	160	143	130
2	201	141	114	99	88	80
3	155	109	88	76	68	62
4	130	91	74	64	57	52
5	114	80	65	56	50	46
6	102	72	59	51	45	41
7	94	66	54	46	41	38
8	87	61	50	43	39	35
9	82	57	47	40	36	33

Table II. (Continued)

New Probe - 1 in. SS pipe: S = 4.7 cpm/mCi/ml
A = 6.8 cpm/mCi
R_r = 7.0 cpm

Detection Limits (μL of D_2O)

^{228}Th mCi	Count Time (min)					
	10	20	30	40	50	60
1	521	364	296	255	228	208
2	316	221	180	155	139	126
3	242	170	138	119	106	97
4	202	142	115	100	89	81
5	177	124	101	87	78	71
6	159	111	91	78	70	64
7	145	102	83	72	64	58
8	134	95	77	67	59	54
9	126	88	72	62	56	51

* All results use the nominal 3 mCi value of ^{228}Th, per Isotopes Products, Inc. Plans for obtaining an accurate measured value are being explored, for future refinement of these results.

TABLE III. Schedule for Monitor Turnover to SRP

Items	Turnover Date
(1) Equipment	
Present hardware	10/5/84
Modification guides*	10/12/84
(2) Maintenance data	
Drawings	10/12/84
Manuals	10/12/84
Purchase orders	10/12/84
(3) Operation aids	
Written procedures	10/12/84
Training session	per H-area date

* New probe geometries may need to be developed, as H-area piping situations are quite varied.

APPENDIX. Definition of Detection Limits

In this work the detection limits are defined as follows: If we have n_b background counts in time t, then a true background count will exceed an alarm level defined as $n_a = n_b + \sigma_b$ with a probability of 16%. Similarly, an assumed valid D_2O detection count n_d (including background) in time t is defined such that $n_d - \sigma_d = n_a$, so that counts below n_a would occur with only 16% probability. The statistical counting uncertainty for any nuclear count n is given by $\sigma = \sqrt{n}$. Thus, we may write

$$n_a = n_b + \sigma_b = n_d - \sigma_d$$

$$n_b + \sqrt{n_b} = n_d - \sqrt{n_d}$$

$$n_d = (\sqrt{n_b} + 1)^2 \qquad \text{(A-1)}$$

The actual counts corresponding to D_2O detected at the limit corresponding to n_d is given by

$$\Delta n = n_d - n_b = (\sqrt{n_b} + 1)^2 - n_b = 2\sqrt{n_b} + 1 \qquad \text{(A-2)}$$

In terms of count rate limit ΔR corresponding to just detected D_2O, we have

$$\Delta R = \Delta n/t = 2\sqrt{R_b/t} + 1/t \qquad \text{(A-3)}$$

where $R_b = n_b/t$ is the background count rate. The D_2O detection limit ΔN_D (μL of D_2O) is calculated as

$$\Delta N_D = R/GS \qquad \text{(A-4)}$$

where G is the gamma source strength (mCi) and S is the probe sensitivity (cpm/μL D_2O/mCi).

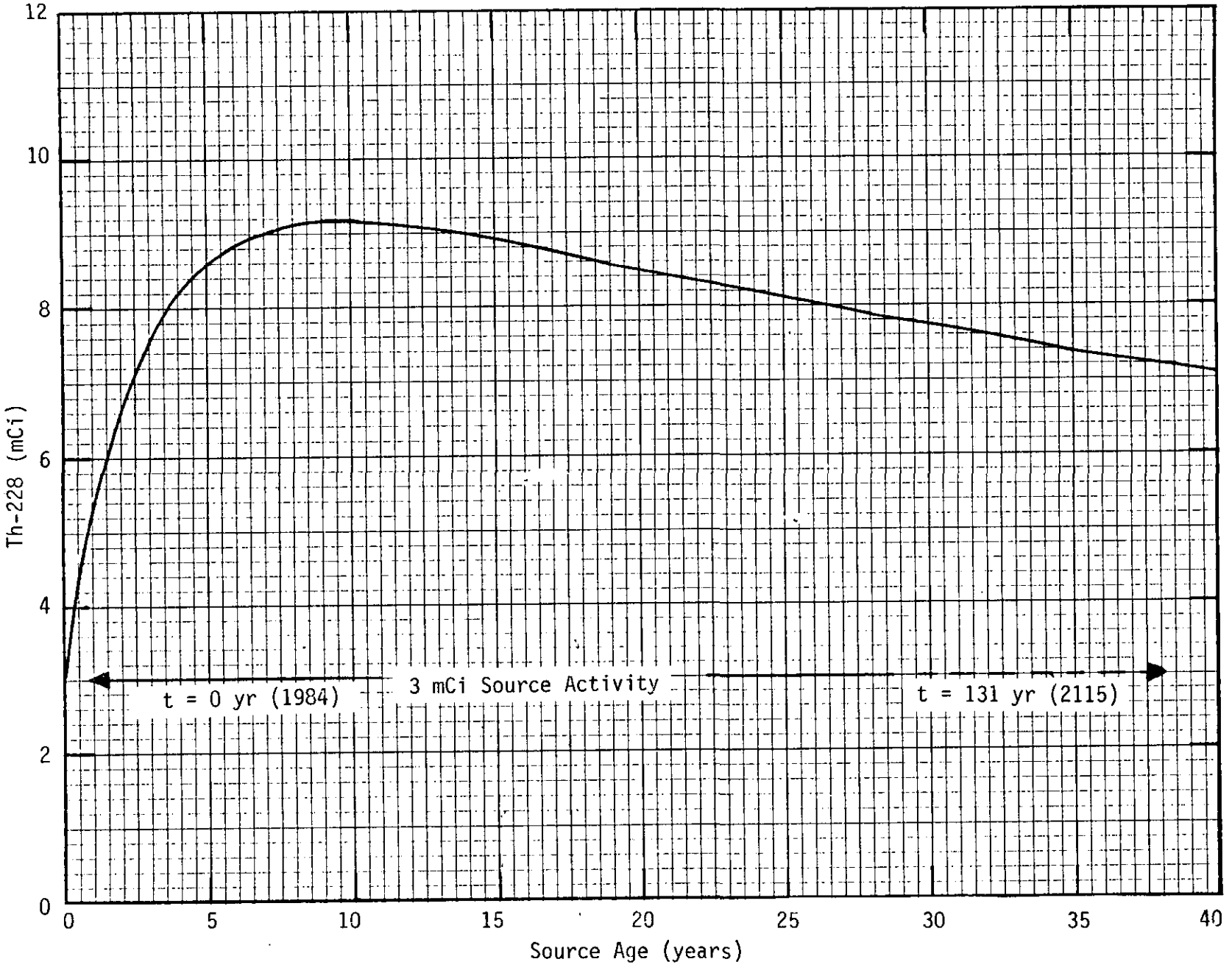

Figure 1. Th-228 Buildup and Decay for Initial 10 mCi/3 mCi U-232/Th-228 Source