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A ${}^2\text{D}(\gamma, n){}^1\text{H}$ TRITIATED-WATER DETECTOR WITH U-232/Th-228 SOURCE

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A paper proposed for publication in
Nuclear Technology

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16 Pages
5 Figures

A $^2\text{D}(\gamma, n)^1\text{H}$ TRITIATED-WATER DETECTOR WITH U-232/Th-228 SOURCE*

Norman P. Baumann and Willard G. Winn

E. I. du Pont de Nemours and Company
Savannah River Laboratory
Aiken, South Carolina 29808**ABSTRACT**

A long-lived gamma source, containing 370 MBq of U-232 (72 yr) and 110 MBq of Th-228 (1.9 yr), was developed as a permanent replacement for neutron-activated Na-24 (15 hr) sources that have been used in a $^2\text{D}(\gamma, n)^1\text{H}$ tritiated-water detector. The Th-228, which will build up to 340 MBq in 10 years and then decay in equilibrium with U-232, emits copious 2614 keV γ -rays for the (γ, n) process. Competing (α, n) backgrounds are reduced by placing the U/Th source in a gold matrix, to provide threshold and Coulomb barriers against the reaction. The detection limits (~ 0.01 mL D_2O) for typical stainless steel pipes (~ 2 cm diameter) measured with the U/Th source are almost as low as those for comparable Na-24 sources, which do not produce (α, n) backgrounds. This minor sensitivity loss can be offset by longer counting times, if necessary. Lower personnel exposure is realized with the new source. Overall, the practical advantages of using a longer-lived source strongly favor the use of U-232/Th-228 instead of Na-24.

* The information contained in this article was developed during the course of work under Contract No. DE-AC09-76SR00001 with the U.S. Department of Energy.

INTRODUCTION

A tritiated-water detector, based on the ${}^2\text{D}(\gamma, n){}^1\text{H}$ reaction, is being used at the Savannah River Plant (SRP) to confirm that process piping presents no release hazard when maintenance operations are under way.⁽¹⁾ The detector actually measures D_2O , which is always a sizeable known fraction of any tritiated water that can exist in these process lines. After 2 years of experience with this monitor, including about 50 individual pipe appraisals, the usefulness of the detector has been well demonstrated. However, experience also showed that monitor operations with neutron-activated Na-24 (15 hr) γ -sources were very cumbersome, because of the source regeneration requirements defined by the short half-life.

The obvious remedy is a γ -source with both a long half-life and a γ -energy that is above the 2222 keV threshold of the ${}^2\text{D}(\gamma, n){}^1\text{H}$ reaction. These requirements were satisfied with a U-232/Th-228 source, which has an equilibrium half-life of 72 yr and emits 2614 keV gamma rays. These gamma rays, which are emitted from equilibrium daughter Tl-208 (2.1 min) of the Th-228 decay chain,⁽²⁾ exceed the above reaction threshold.

U-232/Th-228 SOURCE

The role of the γ -source in the tritiated-water detector is detailed in an earlier report.⁽¹⁾ Briefly, the γ -source is placed in contact with the pipe where tritiated/deuterated water is suspected. The probe, which consists of six He-3 neutron detectors within a Lucite^{®*} moderator, is placed in contact with the pipe, so

* Du Pont trademark for an acrylic resin.

that detection efficiency for neutrons from the ${}^2\text{D}(\gamma, \text{n}){}^1\text{H}$ reaction is optimized. The remainder of the instrument is comprised of standard detector/counting electronics.

The U-232/Th-228 source was developed by Isotope Products, Inc., and contained nominally 370 MBq of U-232 and 110 MBq of Th-228 during this study. Gamma spectroscopy yielded a value of 106 ± 5 MBq for the Th-228 activity. That activity, which is proportional to the 2614 keV γ -activity of the Tl-208, will build up to about 340 MBq in 10 years and eventually decay in equilibrium with U-232 (72 yr), as indicated in Fig. 1. Thus, periodic measurements of the Th-228 γ -activity over the years will yield a refined value for the U-232. Fig. 1 shows that the source will be at least 110 MBq for over 100 years, which provides an obvious advantage over the neutron-activated Na-24 sources used earlier.⁽¹⁾ An added advantage is that personnel exposure from unused low energy gamma rays is less than that for the unused high energy gamma ray of Na-24.

The major concern in the use of the U-232/Th-228 source is the added neutron background from (α, n) reactions within the source.⁽³⁾ For each 2614 keV gamma ray, there are a total of more than 20 alpha particles, with energies ranging up to 10 MeV. The (α, n) reactions are important only in low-Z elements. For heavy elements, high threshold energies and the Coulomb barrier combine to make these reactions negligible. Thus, an obvious solution for the (α, n) problem is to reduce the source material to metal; however,

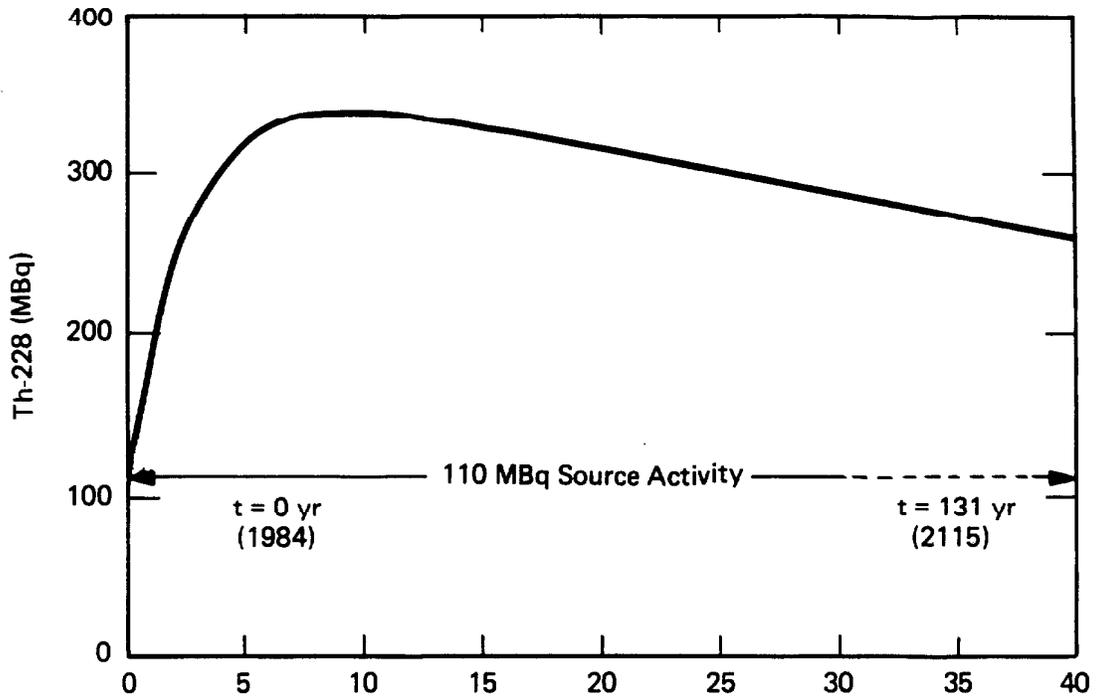


FIG. 1. Th-228 buildup and decay for initial 370 MBq/110 MBq U-232/Th-228 source

this procedure is prohibitively expensive. An alternate approach is to dilute the uranium/thorium salt (or oxide) with finely divided gold or platinum. One practical method is to deposit the salt as a uniform coating on the surface of the thin sheet (~0.03 mm) of gold and then roll it into a compact cylinder. A similar approach was chosen by Isotope Products, Inc. Here, a gold powder (~0.04 mm dia particles) was wetted with chloride salts of uranium and thorium. The powder was then baked and tumbled dry to obtain a good dispersion. Finally, the powder was packed (~75% density) into the 3.2 mm dia x 38 mm long gold-lined cavity of a 6.1 mm dia x 51 mm long stainless steel capsule. All of these operations were performed in a hydrogen atmosphere to minimize oxygen content within the capsule.

Computational estimates were made of the anticipated (α, n) source strength and compared with actual measurements with the source in a calibrated neutron counter. For the estimates, the form of the residual salt was assumed to be uranyl chloride (UO_2Cl_2), and the (α, n) reaction data for oxygen and UO_2 were used.⁽⁴⁾ Although a detailed data set for chlorine was unavailable, it was assumed to be equivalent to that of oxygen, as supported from other data.⁽⁵⁾ Analytical computations were made for source models composed of (1) a homogeneous mixture and (2) stacked parallel slabs with alternating U/Th salts and gold. The thickness of the equivalent gold slab was chosen to give the same surface-to-mass ratio as the spherical particles of the gold powder. The

homogeneous model, which underestimates the effect, predicted a neutron emission rate of 3/sec. The stacked slab model, which geometrically overestimates the effect, yielded a rate of 40/sec.

The measured rate of 33 ± 3 /sec was within the range of the predictions and was demonstrated to be acceptable. This (α, n) background will increase as Th-228 builds up, but somewhat less rapidly than the (γ, n) production. The treatment in the following discussion assumes the two effects are directly proportional, to yield conservative detection limits.

MONITOR PERFORMANCE

Definition of Detection Limit

The monitor performance was appraised by examining the detection limit for different configurations. As in the earlier study, (1) the detection limit is defined as

$$\Delta N_D = \frac{2\sqrt{R_b/t} + 1/t}{SG} \quad (1)$$

where

ΔN_D = detection limit (mL D_2O)

R_b = background count rate (cpm)

t = count time (min)

G = gamma source strength (MBq)

S = detection sensitivity (cpm/mL D_2O /MBq)

When using the U-232/Th-228 source, the background has two components, viz.

$$R_b = R_r + AG \quad (2)$$

where

R_r = room background (cpm)

A = (α, n) sensitivity (cpm/MBq)

R_b depends on the source strength G , which in turn depends on the location of the source relative to the detector. This formalism also can be used for Na-24 sources by setting $A = 0$. Thus, the general form of equation (1) is

$$\Delta N_D = \frac{2 \sqrt{(R_r + AG)/t + 1/t}}{SG} \quad (3)$$

This equation was used to appraise the monitor performance in this study.

Measured Detection Capabilities

Initially, exploratory measurements indicated the optimal approach for using the new source. In essence, it is desirable to minimize ΔN_D by obtaining optimum A and S , per Equation 3. The (α, n) sensitivity A was smallest when the source was mounted on the side of the pipe farthest from the probe, as shown in Fig. 2. The (γ, n) sensitivity S was essentially constant for any source location at the side of a pipe filled with D_2O . For partially filled pipes, the source should be placed on the underside of the pipe, where it is closest to the D_2O and thus can produce the most photo-neutrons, as shown in the figure. This detection configuration is the best because the ratio A/S is minimized.

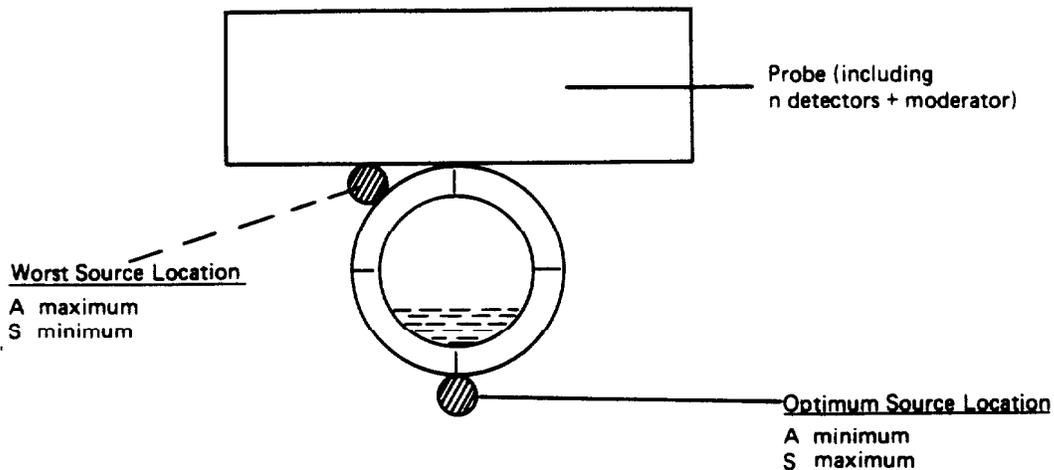


FIG. 2. Optimum detection geometry

For the appraisal measurements, the optimum source/probe configuration was used with pipe sections filled with measured amounts of D_2O . Stainless steel schedule 40 pipes of sizes 1/8 in., 1/4 in., 3/8 in., and 1/2 in. were studied. For each size pipe, count rate measurements were made for room background R_r , total background R_b with the source, and gross count rate R_g with known N_D mL of D_2O in the pipe section. Here, R_g is given by

$$R_g = R_r + AG + SGN_D \quad (4)$$

Using Equations 2 and 4, experimental values of A and S were obtained as

$$A = \frac{R_b - R_r}{G} \quad (5a)$$

$$S = \frac{R_g - R_b}{GN_D} \quad (5b)$$

Measurements were performed for the two probe orientations shown in Fig. 3. Orientation A_1^* offers better detection limits but worse space restraints than orientation B_1^* .

In Fig. 4a, results for S are plotted as $1/\sqrt{S}$ vs. d, where d is the pipe diameter. (Note that the pipe diameter is different from the pipe schedule size.) The approximate linearity of these plots illustrates that S is inversely proportional to the square of an effective source distance from the probe. Similar behavior is also exhibited in the $1/\sqrt{A}$ vs. d plot of Fig. 4b. Here, the data for A correspond to a special A_1^* situation, with an added plastic source holder to enhance the neutron count rate and thus to yield better statistics. The profile of this plot was normalized, using count rate measurements for actual A_1^* and B_1^* cases of A, to yield separate $1/\sqrt{A}$ vs. d predictions for these cases. The resulting estimates of A for the A_1^* and B_1^* cases are reasonable and conservative.

Using the fitted estimates of A and S in Fig. 4 for the A_1^* and B_1^* probe geometries, calculations of ΔN_D as a function of d and G were performed using Equation 3; they are plotted in Fig. 5. In these plots, a room background R_r of 7 cpm and a counting time t of 10 min were used as typical cases. Also plotted are cases for $A = 0$ to illustrate the impact of the (α, n) contribution.

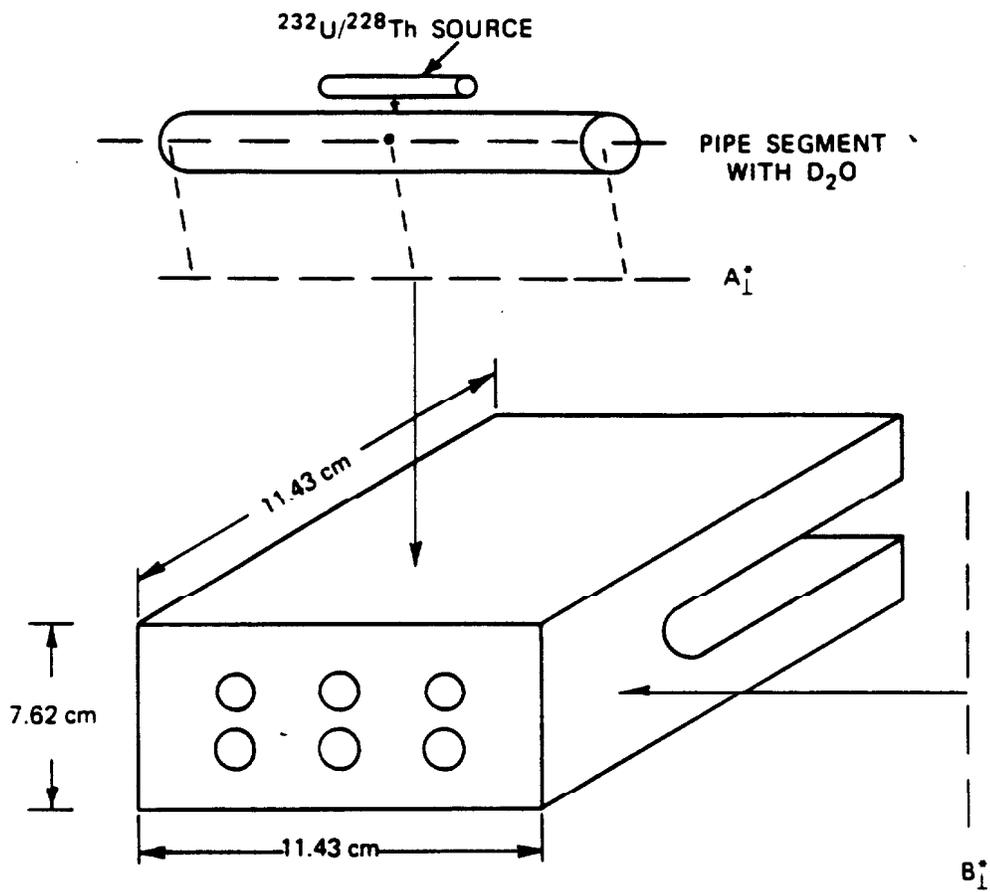


FIG. 3. Detector geometries for stainless steel pipes examined with $^{232}\text{U}/^{228}\text{Th}$ source.

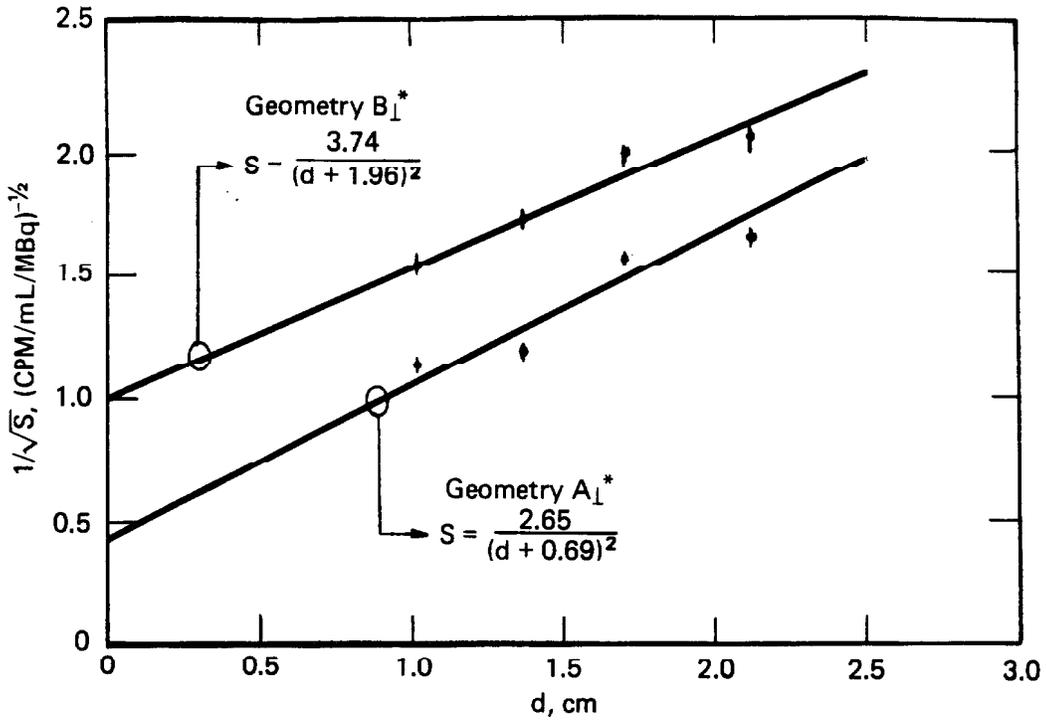


FIG 4a. Functional behavior of S

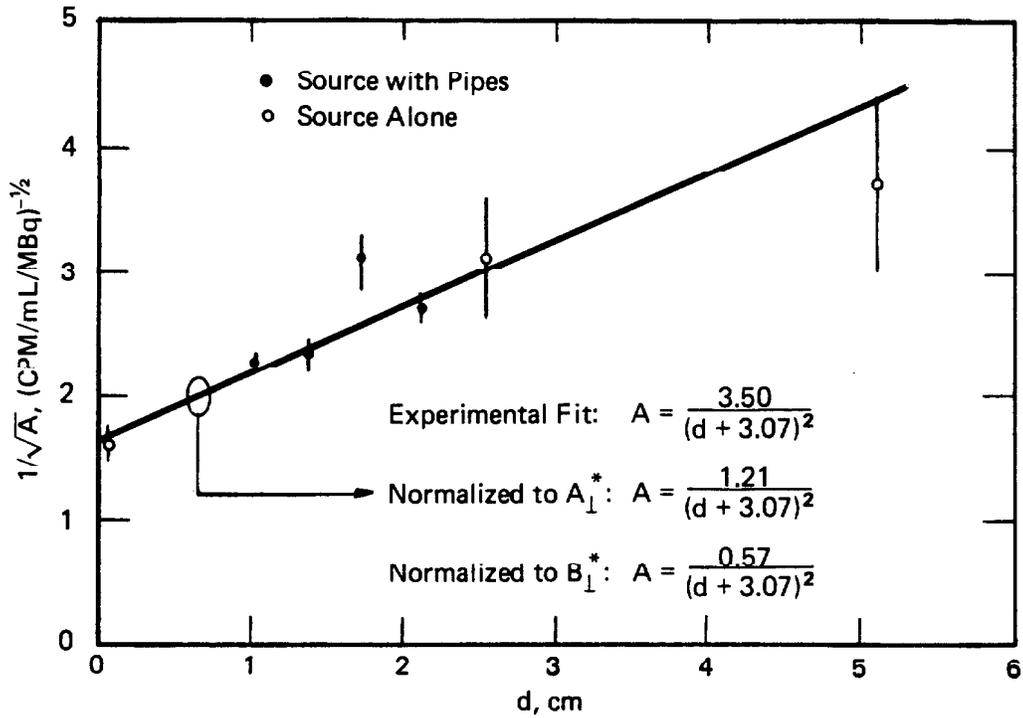


FIG. 4b. Functional behavior of A

Comparison of U-232/Th-228 and Na-24 Sources

A straightforward comparison between equal-strength Na-24 and U-232/Th-228 sources is obtained from Fig. 5. Essentially, the $A = 0$ cases correspond to Na-24, and the others correspond to U-232/Th-228. For this comparison, the (γ, n) production for both sources is taken to be identical. Although the (γ, n) cross section for the 2754 keV gamma of Na-24 is 23% larger than that of the 2614 keV gamma of U-232/Th-228, (6,7) this difference is partially offset by the fact that the Na-24 photoneutrons are 35% more energetic than those of U-232/Th-228 and thus not as readily moderated for detection. Indeed, ANISN calculations(8) of the moderating effect indicate that the detection efficiency for photoneutrons produced by 2754 keV gammas is about 10% lower than for those produced by 2614 keV gammas. The results for the comparison of U-232/Th-228 and Na-24, which focus on the (α, n) effect, also agree qualitatively with direct source comparisons which require geometry, activation/decay, and other normalizations to display this (α, n) effect.

The U-232/Th-228 ($A \neq 0$) and Na-24 ($A = 0$) cases overlap substantially in Fig. 5; thus the (α, n) background effects are minor for the tritiated-water detector. Counting times longer than the 10 min cases in Fig. 5 can be used to improve the detection limit, if necessary.

It should be re-emphasized that these comparisons all assume equal 2614 keV and 2754 keV (γ, n) rates. By contrast, note that the yield of the 2614 keV gamma decay is 36% per Th-228 decay so

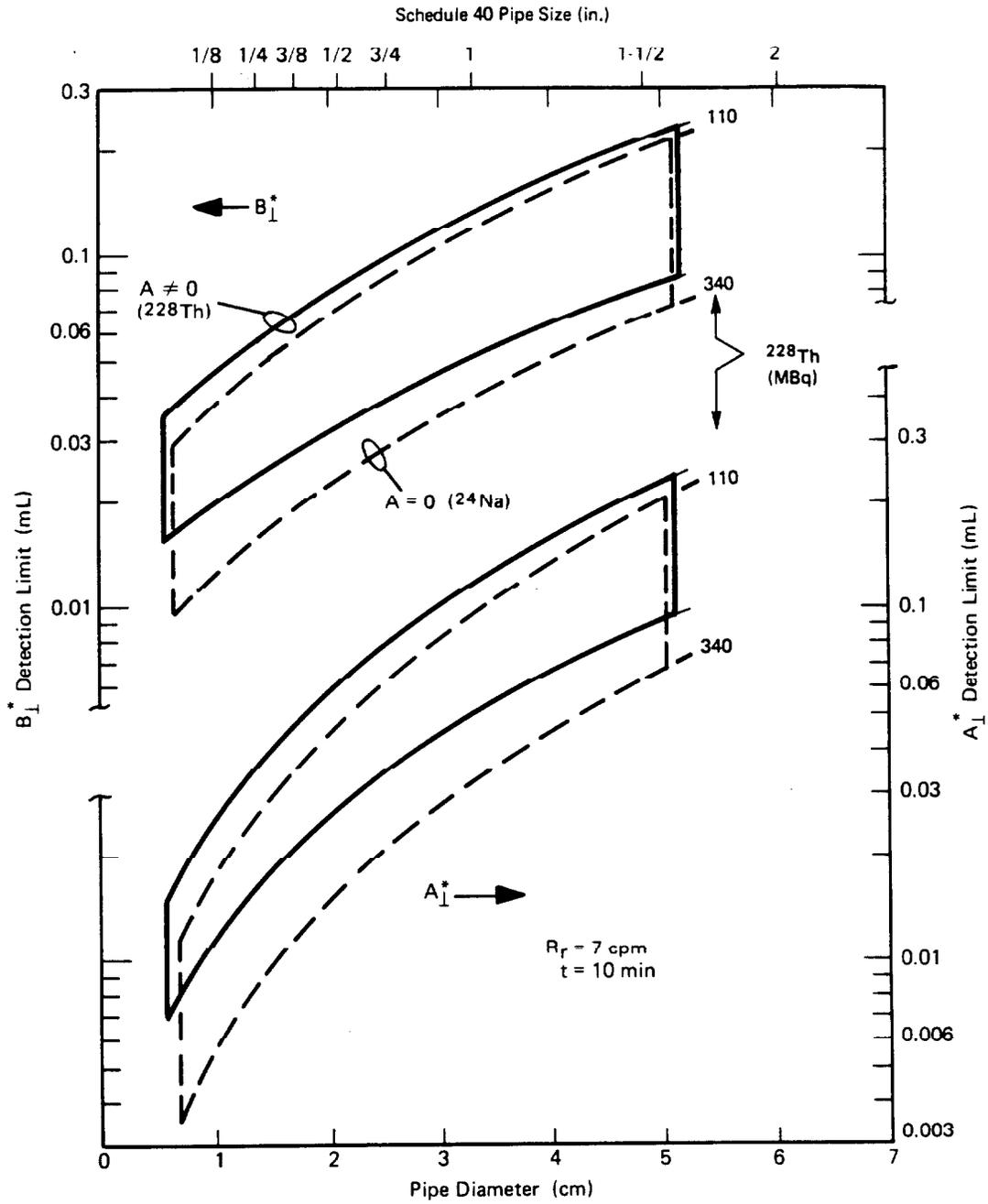


FIG. 5. Detection limit for A_1^* and B_1^* configuration

that almost 3 MBq of Th-228 are required for each MBq of Na-24 in making the above two gamma rates equal. Consequently, the total number of gamma rays in the decay chain for Th-228 is larger than that for Na-24. However, most of these Th gammas have relatively low energy, and their contributions to the "gamma grass" in the neutron detectors showed little impact on neutron detection performance.

CONCLUSIONS

Overall, the detection capabilities of the new U-232/Th-228 source are comparable to those of the Na-24 source. Slight disadvantages resulting from (α ,n) backgrounds can be rectified by longer counting, if necessary.

The main benefit in using the new source is the ease of operation. Elimination of the neutron activation step required for Na-24 sources saves about 24 hours in planning, scheduling, and executing. Significant reduction of this time would be prohibitively expensive. Furthermore, there have been times when no onsite reactor was available for activation service, meaning off-plant arrangements would have been required. With the new U-232/Th-228 source, the monitor can be put in operation in less than 15 min. The long half-life of the U-232/Th-228 source also eliminates the need to record calibration and measurement times, as required for decay corrections when using a Na-24 source. Nevertheless, periodic calibration checks should be made when the monitor is used.

As a result of the good performance and ease of operation with the new source, the monitor is now being used routinely by technicians in plant applications at SRP.

ACKNOWLEDGMENTS

The authors wish to thank R. J. Beleski and R. D. Crawford for assisting in measurements involving plant applications. We wish to thank H. T. Harris for overall coordination of monitor usage in the plant, which encouraged the development of the new source. Finally, we wish to thank Isotope Products, Inc., for developing fabrication methods to produce a source meeting our specifications.

The information contained in this article was developed during the course of work under Contract No. DE-AC09-76SR00001 with the U.S. Department of Energy.

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FIG. 2. Optimum detection geometry.

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FIG. 4a. Functional behavior of S.

FIG. 4b. Functional behavior of A.

FIG. 5. Detection limit for A_1^* and B_1^* configuration.