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A TANTALUM FAST-NEUTRON INTEGRATOR

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ABSTRACT

A very useful fast-neutron detector consists of a slow-neutron sensitive activation element enclosed in a cadmium-clad hydrogenous moderator. When the centrally located element is surrounded by paraffin thickness of about 7 cm, detector response is nearly independent of incident fast-neutron energy throughout the range 0.2 to 20 MeV; the detector is therefore a good flux integrator for a broad region of the neutron spectrum that is of great importance to the accelerator health physicist. Among activation elements so employed, cobalt is widely used for long integration periods, especially at sites where fluxes are low enough to permit continuous occupancy. A single 56-gram cobalt disk can readily measure a flux integral of 1.0×10^7 n/cm^2 when a low-background NaI(Tl) scintillation-crystal gamma-ray spectrometer is used to assay for induced Co^{60} activity.

We describe the newest member of this type of flux integrator, that uses tantalum instead of cobalt, and thereby increases detector sensitivity about one order of magnitude. A 110-gram tantalum disk (2 in. diam by 1/8 in. thick) can easily measure a flux integral of 10^6 n/cm^2, and the minimum detectable integral is almost an order of magnitude lower than this value. The observed activity in tantalum is 115-day Ta-182--a much shorter half-life than 5.26-year Co^{60}; as a consequence, exposure periods for the tantalum flux integrator should be no longer than a few months. Although the time restriction is some disadvantage, the increased sensitivity now permits use of flux integrators in many situations important to radiation protection that were previously beyond the reach of this technique.

I. INTRODUCTION

Fast neutrons can be measured with a detector that consists of a thermal-neutron sensitive activation element enclosed in a cadmium-clad hydrogenous moderator. Such detectors were first used for studying the stray radiation field of high-energy accelerators at UCLRL Berkeley almost 10 years ago.[1] Further development of this technique was reported by Stephens and Smith,[2] who showed a 6-inch diameter moderator to have a response characteristic that was nearly independent of incident-neutron energy throughout the range 0.20 to 20 MeV. Indium was the activation element then used; later, gold was also used, to permit flux integration over many hours. Cobalt, soon added to the list of flux integrators,[3] made possible integration periods of at least a year.

The moderator detectors have seen ever-increasing application in our health physics work at Berkeley, and have come into wide usage at a number of other high-energy accelerator laboratories; such applications are described in the Proceedings from a recent (1965) conference held at the Brookhaven National Laboratory.[4]

Indium and gold are particularly suitable for measuring flux integrals and average flux intensities that exist for short time periods, ranging from a few minutes (indium) to a few days (gold). In addition to providing results that are of immediate use for biological hazard evaluation, these data are always analyzed in terms of the relationship that exists between the neutron field and the specific set of accelerator operating conditions. On the other hand, cobalt is employed for continuous long-term surveillance at a number of locations throughout the project area, and integration periods of 6 months to a year are typical. This integrator data represents a summation of many accelerator operating conditions, and so cannot easily be related to accelerator parameters; however, the data gives directly the integrated fast-neutron exposure received at each irradiation site. Total exposure information is the most important quantity provided by cobalt, and the sensitivity of the detector permits its use in most occupied areas on the project, given an integration period of at least 6 months.

A flux integral of 1.0×10^7 n/cm^2 can be measured easily with the cobalt fast-neutron integrator, when assay for Co^{60} is done with a low-background NaI(Tl) scintillation crystal gamma-ray spectrometer. For example, our S-4 crystal [a 4-in.-diam by 2-in.-thick NaI(Tl) crystal] produces 1.80 c/min in the selected energy band from a single cobalt disk when the detector is exposed to a 10^7 flux integral; in our low-background facility, the corresponding background response is 10.1 c/min. However, measurement of a flux integral smaller than about 5×10^6 n/cm^2 requires longer than an overnight count if the result is to have a statistical standard deviation no larger than ± 10%. An integral of 10^6 is beyond practical reach of a single integrator and the S-4 system. We note that 10^6 n/cm^2 is equivalent to a biological dose at 1-MeV neutron energy of about 33 mrem--a significant exposure. Clearly, a more sensitive long-term flux integrator would be very useful.

II. PRESENT DEVELOPMENT: THE TANTALUM INTEGRATOR

We report the development of a flux integrator of higher sensitivity; this moderator detector uses tantalum instead of cobalt, and thereby provides nearly an order of magnitude increase in sensitivity compared to cobalt. The 115-day half-life of Ta^{182} imposes some restriction on length of an integration period, but intervals of several months are feasible, particularly when some knowledge is available regarding changes in the irradiation rate. If we again use the S-4 spectrometer system for activity assay, a single tantalum disk (2 in. diam by 1/8 in. thick) produces 9.52 c/min in the selected energy band from a 10^7 n/cm^2 flux integral; the corresponding background response is 8.01 c/min. Note that 1.9 c/min will be observed from a 2.0×10^6 n/cm^2 flux integral, and can be measured more easily with tantalum than we could measure a 10^7 n/cm^2 integral with cobalt.

We have a second low-background NaI(Tl) crystal gamma-ray spectrometer, the S-6 system; the detector is an 8-in. -diam by 4-in. -thick scintillator, used primarily to measure very small activities in large-volume samples. With a tantalum disk (2 in. diam by 1/8 in. thick), the 10^7 n/cm^2 flux integral produces 22.3 c/min in the selected energy band, a value to be compared to a BKG response of 33.3 c/min. A detailed comparison of these two systems for counting Ta^{182} and Co^{60} gamma-rays appears in a later section; first, we turn to discussion of integrator calibrations and γ-ray spectral characteristics.

III. CALIBRATION OF TANTALUM INTEGRATOR

All tantalum flux integrator calibrations were done with the same neutron source and the same moderator. The two-piece moderator (the standard design used at LRL Berkeley) is a 6-in. -diam by 6-in. -high right circular cylinder when assembled. The outer jackets of the two parts (halves) are made of cadmium, spun to the correct shape. The moderator material, paraffin, is poured into each shell and the faces are then machined to the desired dimensions. A centrally located depression is milled into one or both moderator faces to receive the activation element. There is a 1-in. overlap of cadmium at the joining plane of the moderator faces.

We used a Pu^{238} Be neutron source for calibration exposures. This source, our number Pu Be 632, has an emission of $8.60 \pm 0.26 \times 10^7$ n/sec, as certified by a National Bureau of Standards calibration. We express the errors in our detector calibration results in terms of the standard deviation; however, such errors are only those that belong with the counting statistics of the gamma-ray spectral analysis. They do not include the $\pm 3\%$ uncertainty in absolute calibration of source emission. This fact must be kept in mind when we compare integrator calibrations that are based on different neutron sources--particularly if these sources are at different laboratories and cannot be directly compared.

We used a low-scatter foam polystyrene assembly for accurate positioning of both source and moderator in all calibration exposures. This assembly was held well above the ground surface to reduce scattering effects still further.

Pertinent data from the calibration exposures are listed on Table I.

Table I. Exposure data for tantalum integrator calibrations.

Calibration run	Tantalum disk description	Exposure conditions	Calibration constant (c/min per 10^7 n/cm^2) S-6 system	S-4 system
1	Ta 2 disk 1/8 in. thick	20-cm distance 244-min exposure 2.50×10^8 n/cm^2	22.3 ± 0.1	9.52 ± 0.07
2	Ta 6 disk 1/8 in. thick	40-cm distance 300-min exposure 7.71×10^7 n/cm^2	22.9 ± 0.2	
3	Ta 8 disk 1/16 in. thick	20-cm distance 90-min exposure 9.23×10^7 n/cm^2	17.0 ± 0.1	
4	Ta 4 and 5 disks 1/4 in. thick	20-cm distance 30-min exposure 3.08×10^7 n/cm^2	25.4 ± 0.2	

Source-to-moderator distances are center-to-center distances between the two objects during exposures. All values for calibration constants listed here have been determined experimentally. We adopt the value 22.3 c/min per 10^7 n/cm^2 to be the tantalum integrator calibration constant for the S-6 system, when a 2-in.-diam by 1/8-in.-thick disk is used in the moderator; this result is obtained from Exposure 1 (calibration run 1). Exposure 2 was done to determine whether this result could be in error because of the relatively short source-to-moderator distance used in Exposure 1. (Previous work by McCaslin had shown that calibrations performed at a 20-cm distance were valid for gold and indium foils of 1 in. diam.[5]) A comparison of the two values shows that they agree within two standard deviations; this is not conclusive evidence that the values are different, and so we consider the 20-cm distance (Exposure 1) to give valid calibration constants.

We observe the effect of different disk thicknesses in the results from Exposures 3 and 4. The 1/8 in. thickness provides a significant sensitivity increase compared to the 1/16-in.-thick disk. Although a 1/4-in.-thick disk shows some sensitivity increase compared to a 1/8 in. thickness, it is doubtful that this represents a worthwhile increase. A much greater sensitivity increase can be achieved by exposing a pair of 1/8-in.-thick disks in separate moderators; the disks can be counted together, to provide very nearly a factor of 2 increase.

Exposure 1 also provided data for the calibration constant of the S-4 system; the value is 9.52 c/min per 10^7 n/cm^2 for the 1/8-in. -thick disk. None of the other calibration exposure disks were counted with S-4; we have chosen not to enter S-4 values for these disks, because the values were not experimentally determined.

IV. γ-RAY SPECTRUM OF ACTIVATED TANTALUM DISKS

Gamma-ray spectral analysis is usually done with a NaI(Tl) scintillation crystal; in fact, only through use of such a high-sensitivity detector can we measure small flux integrals. The lower curve on Fig. 1 portrays the spectrum from calibration disk Ta 2, determined with the S-6 system. We select the energy interval from 1.02 to 1.28 MeV (channels 103 through 129, inclusive) to indicate Ta^{182} activity; vertical bars on the curve mark these limits. Note that two rather broad peaks are evident; however, the saddle between them is very shallow because spectrometer resolution is not good enough to provide clear separation of closely spaced gamma-ray lines. Better separation can be seen in a report by Heath,[6] and was achieved by use of a NaI(Tl) crystal of higher-resolution. However, the NaI(Tl) single-crystal spectrometer is unsuitable for studying details of so complex a gamma-ray spectrum, as will become evident from the following discussion.

It is very important that only Ta^{182} activity contribute counts to the selected energy interval; the truth of this statement must be demonstrated before tantalum integrator data can be of much value. Because of gamma-ray spectral complexity and poor spectrometer resolution, NaI(Tl) crystal data could decide the question only by showing that a half-life appropriate to Ta^{182} were observed during repetitive analysis of an activated disk. Such a series of measurements might span many months, in view of the 115-day Ta^{182} half-life. We have chosen a quicker and more direct method to prove the point.

Given sufficient sample activity, the Ge(Li) crystal is now the logical choice for gamma-ray spectrum investigation. Henck et al. have made such a study of Ta^{182}-decay;[7] his values for gamma-ray energies and relative intensities are used in Table II. There are four prominent gamma rays which have energies within the selected interval; these and several weaker ones all contribute counts that we interpret to mean Ta^{182} activity.

We have examined gamma-ray spectra from tantalum disks with a Ge(Li) crystal, a detector 2 by 3 by 1 cm thick. Detector-output pulses are coupled through low-noise amplifier stages to a 1600-channel pulse-height analyzer. Energy calibration of the system for this study is very nearly 1 keV/ channel, meaning that gamma-ray energy is approximately equal to channel number. A slight nonlinearity exists in the energy-vs-channel-number relationship; its nature has been investigated with known-energy gamma-ray lines, so that we can assign gamma-ray-energy values to within about 1 keV simply by noting peak positions on plotted spectra and applying a single correction. Energy values appearing in the last column of Table I were obtained in this manner from the data used to sketch Fig. 2.

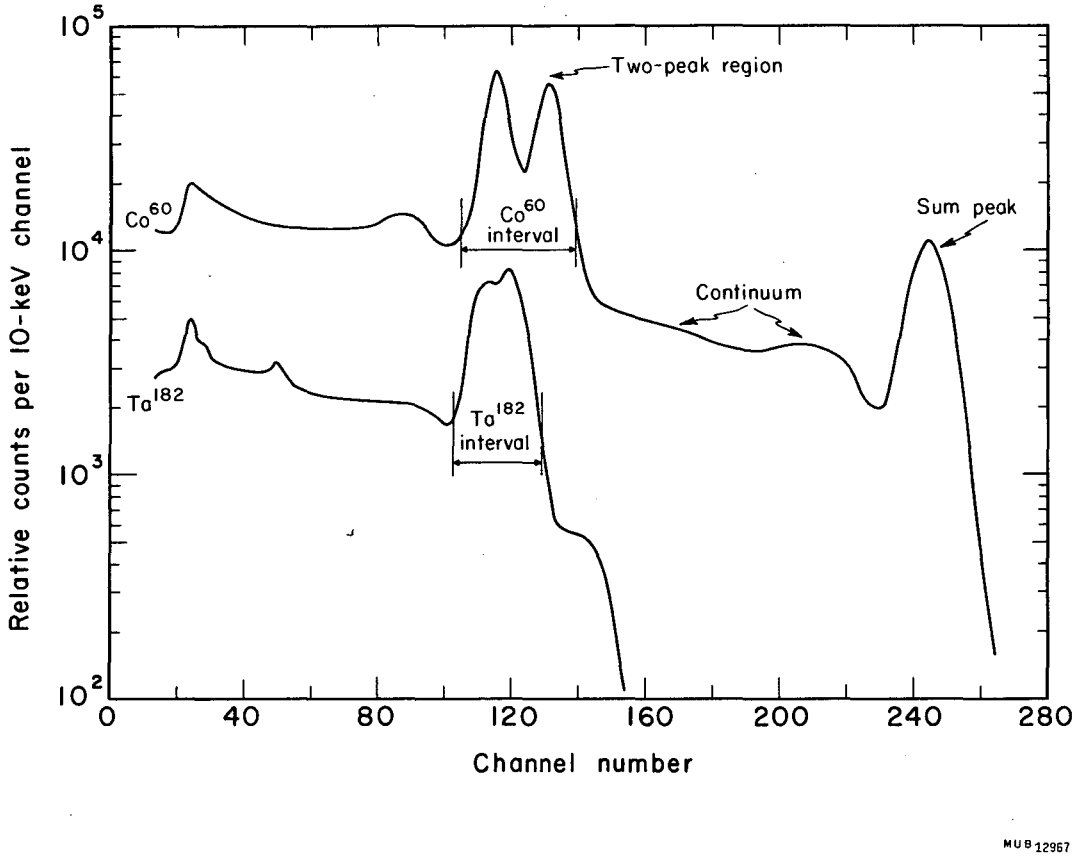

Fig. 1. NaI(Tl) scintillation crystal gamma-ray spectra of Co^{60} and Ta^{182} observed in activated disks.

Table II. Information concerning gamma-ray emission that accompanies the decay of Ta^{182} and Co^{60}.

Tantalum integrator

Active element: tantalum disk, 2 in. diam by 1/8 in. thick, weight 110 grams
Reaction: $Ta^{181}(n, \gamma) Ta^{182}$; 115 day half life

Gamma rays > 1.100 MeV			
Henck et al.[a]		Present work	
1.122 MeV	(100)	1.122	MeV
1.158	(3)	1.158	
1.189	(44)	1.190	
1.222	(72)	1.222	
1.230	(36)	1.231	
1.258	(5)	1.258	
1.275	(2)	1.275	
1.289	(4)	1.290	
1.343	(0.8)	1.343	
1.374	(0.7)	1.374	

None in coincidence

Values in parentheses are relative abundances.

Cobalt integrator

Active element: cobalt disk, 2 in. diam by 1/8 in. thick, weight 56 grams
Reaction: $Co^{59}(n, \gamma) Co^{60}$; 5.26 year half life

Gamma rays: 1.1728 MeV (100%)
1.1325 MeV (100%)
Always in coincidence
Values in parentheses are absolute abundances per decay

a. Values taken from Ref. 7.

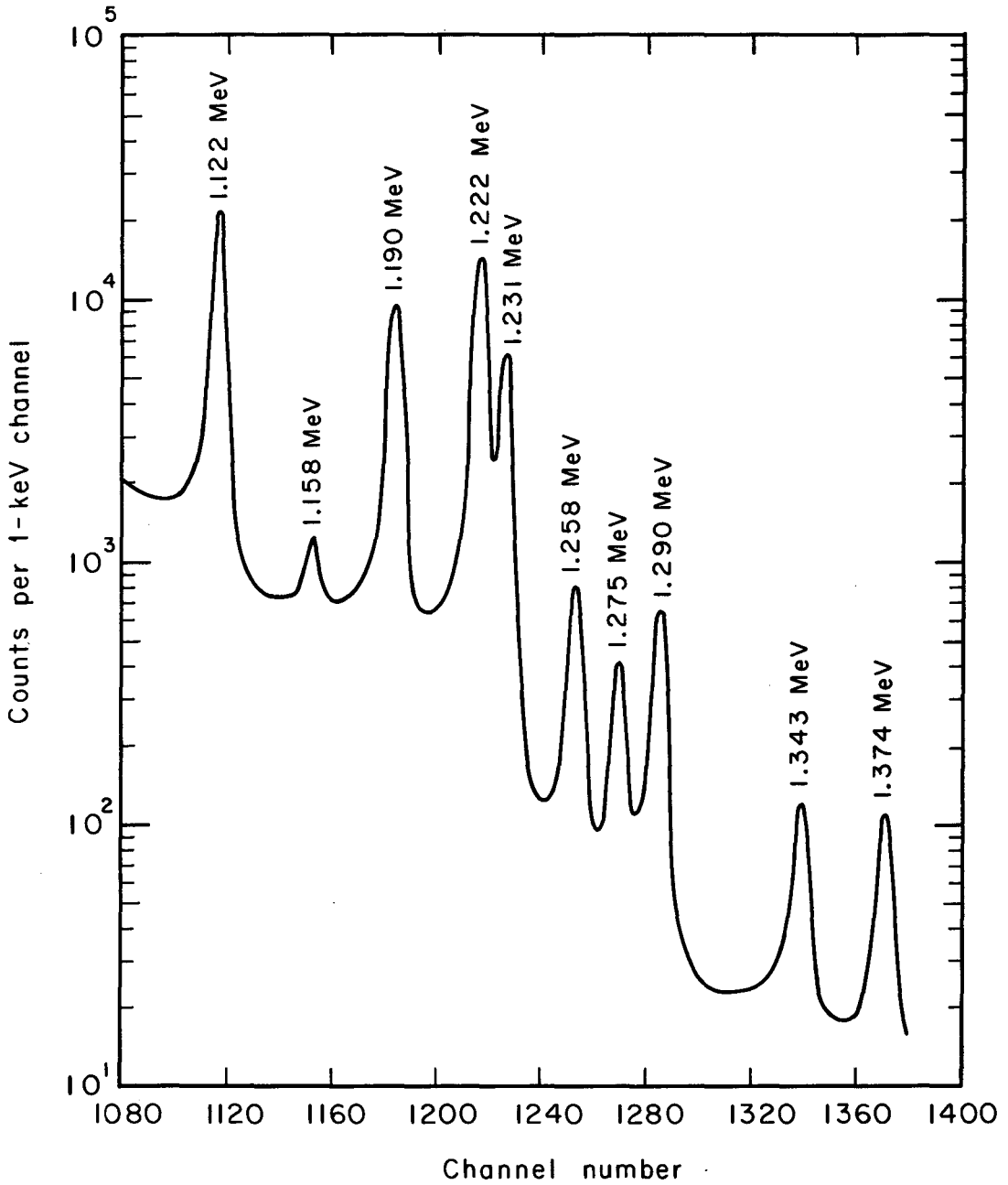

Fig. 2. Ge(Li) semiconductor crystal gamma-ray spectrum of Ta^{182}, showing only the energy interval of interest. (1080 to 1400 keV)

Tantalum disks were activated in two ways for the Ge(Li) study:

1. Disk in moderator exposed to the calibration neutron source, Pu^{238} Be 632. From the Pu Be exposure, we verified that only Ta^{182} appeared in the Ge(Li) spectrum; we could therefore use NaI(Tl) data taken shortly after an exposure to generate accurate detector-calibration constants.
2. Disk in moderator exposed in an unshielded site near a thick target at the Bevatron, located so that a significant fraction of the flux consisted of >50-MeV particles. From the Bevatron exposure, we could learn what competing activities (if any) are produced when a tantalum integrator is exposed to high-energy particles.

Figure 2 shows a free-hand version of the important region of a Ge(Li) crystal spectrum (1080 to 1400 keV) from this disk; the spectrum was acquired in 600 minutes of counting time, that began 12 hours after the end of a 60-hour exposure. All peaks are identified as Ta^{182}, by both position and relative intensity. A shorter Ge(Li) run taken 1 hour after exposure end failed to reveal any significant competing activities. Because of poorer counting statistics, the short run does not present as clearly defined a spectrum as that shown in Fig. 2. We omit a plot of the short-run spectrum for this reason, although its data is quite important to the point under consideration.

We conclude that competing activities are unlikely to interfere with Ta^{182}-activity measurement, even when an integrator disk is counted shortly after exposure to a significant flux of >50-MeV particles. The Ge(Li) crystal spectrometer is seen to be extremely useful in providing quick and conclusive evidence to prove this point. As a result, Ta^{182}-activity assay can almost always be done in complete confidence with the high-sensitivity NaI(Tl) scintillation spectrometer. A single exception might be noted: direct exposure of an integrator to a high-energy beam. Here, choice of the proper calibration constant for the integrator is likely to be a much more serious problem than correction for the competing activities.

As a final comment, we mention that our survey of the known decay radiations from high-energy spallation reactions on Ta^{181} does not predict serious interference from such isotopes, provided we use the high-energy Ta^{182} peaks for assay. If low-energy Ta^{182} peaks (100 to 300 keV region) were used, competing activities might pose a serious problem, although the Bevatron exposure did not give clear indication that such is the case.

V. COMPARISON OF THE S-4 AND S-6 SPECTROMETER SYSTEMS

We use both the S-4 and S-6 spectrometer systems to count cobalt and tantalum flux integrator disks. Important descriptive information concerning these systems is listed in Table III; we include calibration constants for both cobalt and tantalum along with the corresponding BKG response values.

It is instructive to study performance of the two systems for detecting Co^{60} and Ta^{182} γ-rays, and so to determine whether the large S-6 crystal provides any advantage when small disk samples are counted. Such a study involves relationships among these parameters: flux integral magnitude,

Table III. Important characteristics of the S-4 and S-6 spectrometer systems.

S-4 spectrometer system	
Detector:	NaI(Tl) crystal, 4 in. diam by 2 in. thick, in low-activity Harshaw "Matched Window" assembly, with a single 3-in.-diam phototube (Dumont type 6363).
Shield:	Small-volume Pb shield in low-background counting facility; low-activity Pb from St. Joseph Mine, Missouri.
BKG:	141.6 c/min in interval 0.1 to 2.1 MeV 10.1 c/min in Co^{60} counting interval 8.01 c/min in Ta^{182} counting interval
Calibration constants (2-in.-diam by 1/8-in.-thick disk):	
	1.80 c/min for cobalt, from 1.0×10^{7} n/cm^2 9.52 c/min for tantalum, from 1.0×10^{7} n/cm^2
S-6 spectrometer system	
Detector:	NaI(Tl) crystal, 8 in. diam by 4 in. thick, in low-activity stainless steel assembly with fuzed-quartz optical window, coupled to single 5-in.-diam phototube, low-activity EMI type 9530-Q.
Shield:	Small-volume Pb shield in low-background counting facility; low-activity Pb from St. Joseph Mine, Missouri.
BKG:	395 c/min in interval 0.1 to 2.1 MeV 53.0 c/min in Co^{60} counting interval 33.3 c/min in Ta^{182} counting interval
Calibration constants (2-in.-diam by 1/8-in.-thick disk):	
	3.80 c/min for cobalt, from 1.0×10^{7} n/cm^2 22.3 c/min for tantalum, from 1.0×10^{7} n/cm^2

count time, calibration constant, precision of result, and BKG response. BKG response values are here taken to be constants, and are the values appropriate to the detectors as described in Table III.

Meaningful comparisons of the two systems can be made in the following way. We specify the precision required for measurement of a flux integral, and then investigate the relationship between flux integral magnitude and counting time needed to achieve this precision. We choose a standard deviation of ± 10% to be the required precision. Table IV lists a range of flux integral values, along with the count times required for both spectrometer systems to produce results from tantalum and cobalt; Fig. 3 portrays these relationships in graphical form. It is clearly an advantage when the given precision for a particular flux integral can be achieved in a shorter counting time; we adopt this criterion to evaluate the systems. In all cases, integration time is assumed short in comparison to the half-life of the radioisotope.

With cobalt, the S-6 system is slightly less effective for determining flux integrals $< 2 \times 10^7$ n/cm^2 in comparison to the S-4 system; the requirement of about 20% longer counting time is approached as the integral becomes smaller. A major cause for the relatively poor S-6 performance is the mode of Co^{60} decay. As shown on Table I, the two gamma rays are always in coincidence. We must use a very short sample-to-detector distance during counting, and many of the coincident pairs enter the crystal to interact; furthermore, many of these interactions deposit enough energy in the large crystal to produce pulse heights that lie beyond the two-peak interval used for Co^{60} assay. A number of such events appear in the sum peak; in fact, if we include the sum peak area along with the two-peak area, the S-6 system performs about as well as the S-4 system for measuring small flux integrals. The upper curve on Fig. 1 is an S-6 spectrum from a cobalt disk, showing the features discussed here. However, the intervening continuum cannot be salvaged because of an unfavorable BKG to SIGNAL ratio. Thus, an apparent large increase in detection efficiency provided by the S-6 crystal cannot always be fully realized when coincident gamma-rays are studied with a single crystal.

In contrast, we see the clear advantage of S-6 for measuring all flux integrals determined by tantalum. Here, no gamma rays with energies greater than 1 MeV are in coincidence, as indicated in Table I. Although some gamma rays of interest are accompanied by coincident low-energy gamma rays, such coincident events will usually remain within the rather broad energy band accepted for Ta^{182} assay. Even for very small integrals, about 35% less analysis time is required for S-6 than for S-4.

We emphasize the fact that our success in using the large S-6 crystal for small flux integrals is due mainly to the very low BKG environment provided for this detector. The S-6 crystal size is not well-matched to the size of the small disk samples. A low-activity 4-in.-diam by 4-in.-thick crystal would probably give better performance than S-6 for small integrals; a pair of crystals like S-4, counting the same disk with summed output signals, should also improve upon the S-6 performance for small integrals measured with tantalum.

Table IV. Data used for comparing performance of S-4 and S-6 spectrometer systems for counting Co^{60} and Ta^{182} in small disk samples.

Count time for standard deviation = ± 10%.

	COBALT INTEGRATOR		TANTALUM INTEGRATOR	
Flux integral (n/cm^2)	S-4 system (min)	S-6 system (min)	S-4 system (min)	S-6 system (min)
1×10^{10}	0.0558	0.0267	----	----
5×10^{9}	0.112	0.0542	0.0210	----
2×10^{9}	0.285	0.140	0.0527	0.0223
1×10^{9}	0.590	0.301	0.106	0.0454
5×10^{8}	1.23	0.673	0.214	0.0923
2×10^{8}	3.54	2.22	0.548	0.238
1×10^{8}	8.7	6.3	1.15	0.50
5×10^{7}	23.6	19.9	2.45	1.16
2×10^{7}	105	104	7.48	3.88
1×10^{7}	367	395	19.4	11.1
5×10^{6}	1 360	1 520	56.4	35.3
2×10^{6}	8 070	9 270	274	186
1×10^{6}	31 700	37 100	993	710
5×10^{5}	126 000	147 500	3 750	2 730
2×10^{5}	very long	very long	22 700	16 600
1×10^{5}	very long	very long	90 000	66 700

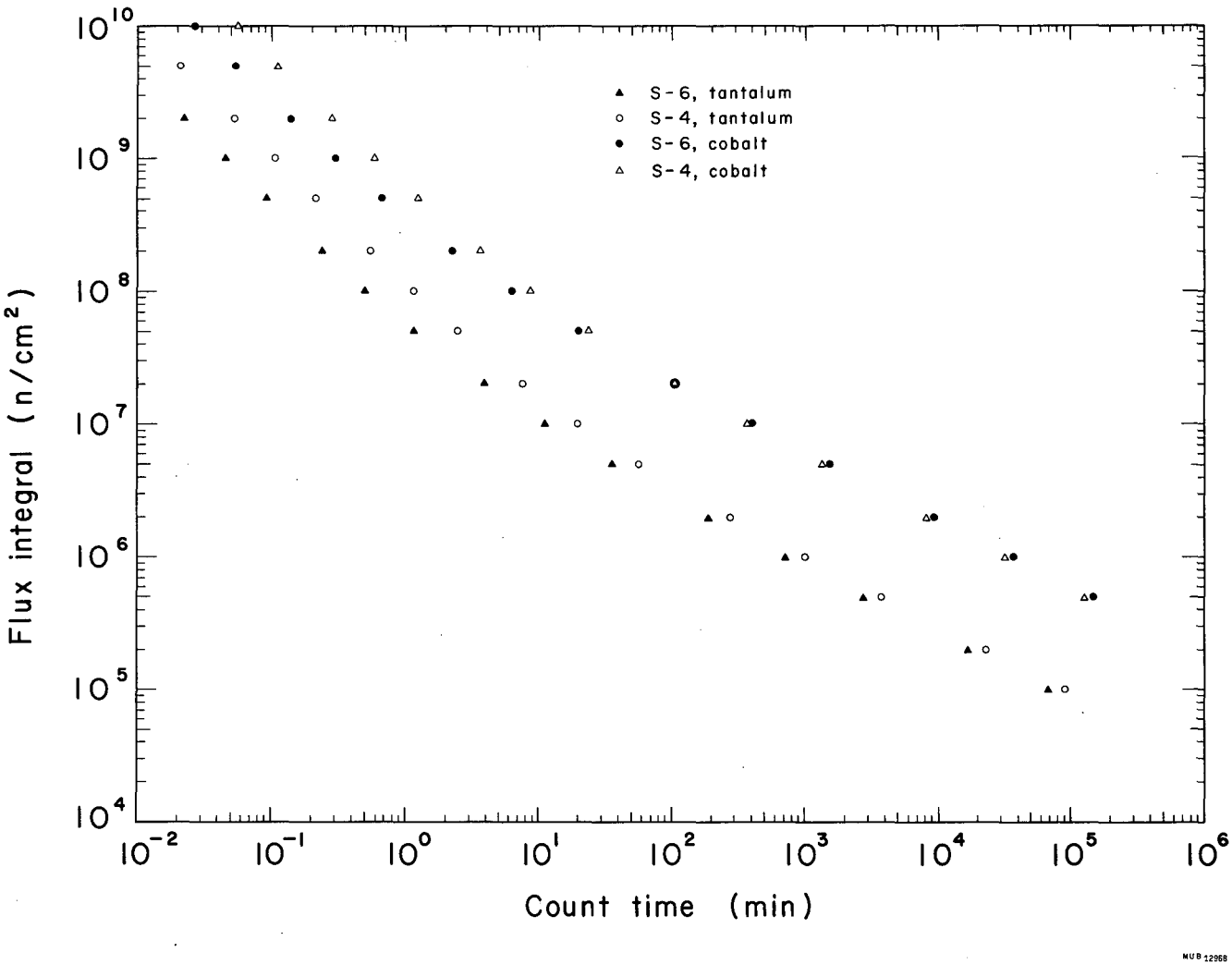

Fig. 3. Comparison of performance of S-4 and S-6 spectrometer systems for counting Co^{60} and Ta^{182} in small disk samples.

For large flux integrals, the S-6 system shows clear advantage when counting either cobalt or tantalum. This advantage becomes simply the ratio of calibration constants for the two systems: the values are 2.33 for tantalum and 2.11 for cobalt. A practical interpretation of these ratios is: compared to S-6, S-4 requires a counting interval 2.33 times as long for tantalum, and 2.11 times as long for cobalt, to produce results that have the same precision. It makes little real difference which system is used when we require only the ± 10% precision from large flux integrals. However, if we desire ± 3% precision, counting time increases about a factor of 10 compared to values listed in Table IV, and ± 1% precision requires a factor of 100 increase in counting time. Differences in counting times then become quite significant, and the advantage of S-6 is evident.

VI. CONCLUSION

We have described a new development in the activation-element type of fast-neutron flux integrator: the use of tantalum as the activation element. We can measure a flux integral of 1.0×10^6 n/cm^2 as easily with tantalum as we can measure a flux integral of 1.0×10^7 n/cm^2 with cobalt, thus providing about one order of magnitude increase in sensitivity. No activities other than the desired Ta^{182} are observed in the selected γ-ray spectral interval, even when the irradiation flux contains a significant high-energy (>50 MeV/nucleon) component; we conclude that competing activities will not cause errors in data interpretation from normal exposure circumstances of this detector. Although the 115-day half life of Ta^{182} imposes some constraints on the length of integration periods, the ability to measure flux integrals in the range 3 to 10×10^5 n/cm^2 (corresponding to a biological dose range of about 10 to 30 mrem at 1-MeV neutron energy) should make this integrator a very useful tool for health physics purposes.

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