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DE88 003166

TITLE A CONFIRMATORY MEASUREMENT TECHNIQUE FOR HEU

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SUBMITTED TO American Nuclear Society
Third International Conference
on Facility Operations - Safeguards Interface
San Diego, California
November 29-December 3, 1987
(FULL PAPER)

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A CONFIRMATORY MEASUREMENT TECHNIQUE FOR HEU

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ABSTRACT

Precise measurements of the special nuclear material (SNM) in an item can be used to confirm that the item has not been tampered with. These measurements do not require a highly accurate calibration, but they should be based on an attribute that is unique to the SNM. We describe an instrument that performs gamma-ray measurements at three energies: 185.7 keV, 1001 keV, and 2614 keV. This instrument collects data for 100 s from shipping containers ([illegible] barrels). These measurements help to distinguish the issue of material control—has any material been diverted?—from the issue of measurement control—is there a measurement bias?

I. INTRODUCTION

Present DOE regulations require that a receiver of a special nuclear material (SNM) shipment perform an input accountability measurement within 10 calendar days of receipt of the shipment. [illegible] those cases for which this measurement is not possible, a confirmatory measurement may be used on an interim basis [illegible] [illegible] for. Some [illegible] specific confirmatory measurement [illegible] [illegible] they are chosen and how they might be better used is under way.[1] [illegible] measurements can be [illegible] the three roles of safeguarding SNM [illegible] [illegible]

[illegible]

[illegible]

[illegible]

(3) to protect the financial interests of the entities that operate the facilities that handle and process SNM.

The first two objectives can be satisfied by precise measurements of the SNM in an item, and the third objective requires accurate measurements of the quantity of SNM in the item.

The causes for shipper receiver (S/R) differences can be separated into two categories: either the item has been altered, or it has not been measured correctly. We shall define these two issues as material control and measurement control. Material control can be accomplished with precise measurements of the SNM, while measurement control requires accurate and precise measurements of the SNM. It has been proposed that a confirmatory measurement [illegible] deal with the material control issue of whether SNM has been diverted or lost. Thus, [illegible] this definition of a confirmatory measurement, we can report the result in counts per [illegible]. This measurement implies that [illegible] [illegible] must be used by both the shipper [illegible] receiver to confirm a shipment of SNM. [illegible] confirmatory measurements [illegible] [illegible] demonstrate that [illegible] SNM [illegible] that the transfer was successful. [illegible] measurement [illegible] between the shipper [illegible] [illegible] accountability [illegible] separate measurement [illegible] [illegible] be addressed at a later [illegible] through this measurement [illegible] [illegible] is peripheral [illegible]

[illegible]

[illegible]

* [illegible]

(2) Short count times with a minimum of sample preparation.

(3) Minimal cost for capital equipment and the personnel to operate it.

(4) Confirmatory measurement difficult to deceive with a bogus item.

If the confirmatory measurement is also used to deduce the sample mass, a nominal measurement accuracy of 10% (1σ) is reasonable. (Better accuracy is not precluded.)

The primary objective of this instrument is to confirm that SNM transferred between facilities has not been lost o diverted. It was immediately obvious that to satisfy requirement (2) above, a confirmatory measurement on the unopened shipping containers would be preferable to measuring the two or three inner containers that reside within each shipping container.

II. THE SAMPLES

These measurements will be performed on sealed shipping containers. The preservation of the original tamper indication will be useful if measurement biases result when the material is opened and analyzed before being put into the process. In addition, there is less chance of contamination or spills if the outside containers are not opened. Finally, considerably less manpower and facilities will be required to safely handle the items.

The outer container is usually a 208-ℓ barrel. The inside geometry of these barrels can vary widely. But in general terms, criticality safety nearly always requires that these barrels have a 5- or 6-in.-diam pipe in their centers. The pipe is surrounded by low Z, low-density packing material (which may be somewhat hydrogenous), and it holds two 2 ℓ or 4 ℓ bottles contained in plastic bags. The contents of the bottles are pure highly enriched uranium (HEU) in the form of oxide, metal, or uranyl nitrate hexahydrite (UNH).

III. THE FOUR GAMMA TECHNIQUE

Despite the large quantities of uranium involved, the passive neutron signal is too weak to provide a precise signal. The potential active neutron techniques are costly and have difficulty penetrating to the center of large quantities of pure HEU. And to compound the issue of sample penetrability, some of the HEU under consideration contains large quantities of water (UNH) that vary during storage or shipment. Consequently, we developed a gamma-ray based approach.

The four-gamma technique consists of a weight measurement and the detection of four gamma rays of different energies. The 185.7-keV gamma ray occurs when ^{235}U decays. In many of the items considered here, the uranium-bearing material is infinitely thick to the 185.7 keV gamma ray. The 1001-keV gamma ray comes from a daughter of the ^{238}U. In freshly processed uranium, this gamma ray may not be in equilibrium. For most of the items considered here, the samples are not infinitely thick (33 cm for oxide/UNH and 4.2 cm for metal). For nearly all of the items under consideration, this signal correlates with ^{238}U mass. In addition, the large quantities of HEU in these samples provide good precision in the 1001-keV gamma-ray measurement. Combining the 185.7- and 1001-keV gamma-ray signals can be useful in estimating the sample enrichment or the sample self-absorption. The 2614-keV gamma ray is the result of the decay of a ^{232}U daughter. The ^{232}U in enriched uranium results from inserting reactor returns into the enrichment plant feed stream. The 2614-keV gamma ray is not in equilibrium with the ^{232}U. In addition, various quantities of reactor returns are in various batches of HEU. Therefore, the 2614 does not correlate with the amount of ^{235}U or ^{238}U, and its intensity can not be easily predicted. Consequently, the 2614 can be used as a tag or unique identifier for each sample.

The four-gamma instrument will also look for a peak in the 350- to 450-keV region. If a peak is found, it suggests that the shipping container might contain plutonium. For most uranium facilities, the presence of plutonium in a shipping container is a problem that occurs rarely, if at all, and that is best addressed by timely return of the unopened shipping container.

IV. THE PROTOTYPE INSTRUMENT

In this section we describe a prototype system for in-plant use. Four low-resolution gamma-ray detectors each containing [illegible]-cm americium-seeded NaI(Tl) crystals will be stacked vertically to view the shipping container. Figure 1 shows the detectors in the detector shields viewing the potential shipping container geometries. The vertical spacing between centers of the NaI(Tl) crystals is 51 cm. Each detector will count the four gamma rays mentioned in the previous section. This yields [illegible] independent count rates for each sample.

The three potential shipping containers are 111-, 208-, and 416-ℓ drums. The four detectors are spaced and collimated to give a uniform response with respect to the sample height. The shipping containers will be rotated during the data acquisition to minimize the effects of off-center positioning of the HEU inside the containers. The instrument's response to variations in sample positioning are discussed [illegible].

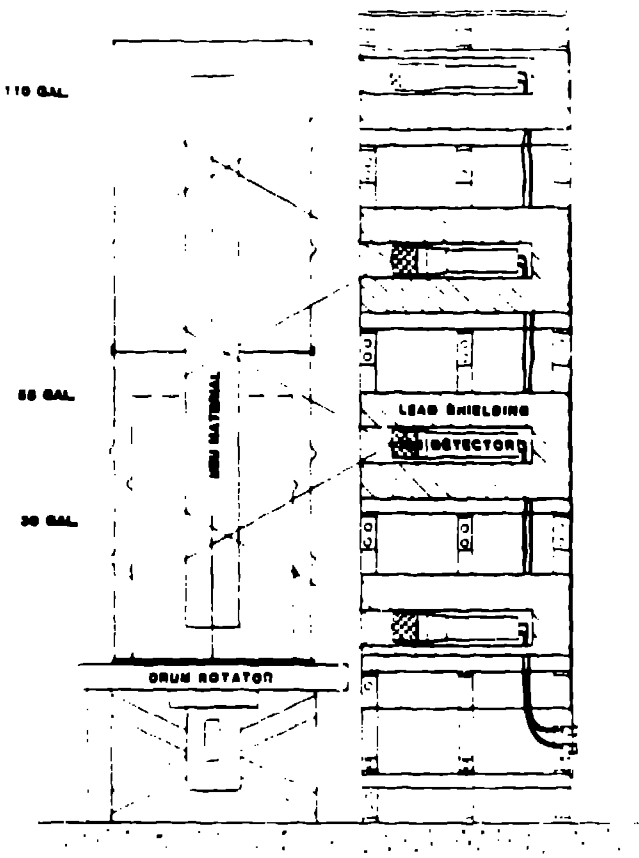

Fig. 1. The prototype HEU confirmatory measurement counter. Four shielded low-resolution gamma ray detectors view the three possible shipping containers: 30-, 55-, and 110-gal. drums. The inner pipe that defines the possible location of the HEU is also shown.

V. THE ANALYSIS

Standard techniques using peak and background regions of interest will determine the net peak areas. These data are normalized by the net peak area of the americium seed pulse. Results from a background run are subtracted; the background corrected normalized net peak areas, A(i), will be the reported results in the units of counts per second. By using three statistical tests for consistency, we will compare results A(i) and B(i) obtained from two instruments.

The first comparison is performed on an individual basis between the 16 pairs of results. The two uncertainties in each pair will be compared to make sure they are similar. If they are, the uncertainties will be combined to form an estimated sigma, s(i). Next, the difference between the two results will be standardized,

$$z(i) = [A(i) - B(i)]/s(i) \quad ,$$

for i = 1, 2, ..., and 16. The z(i) are compared with two thresholds: a warning threshold that indicates the data might be inconsistent and an action threshold that indicates the two results strongly disagree. The thresholds are obtained from tabulated values of the standardized normal distribution.

The second comparison computes a reduced chi-square from the 16 results, χ^2:

$$\chi^2 = \left[\sum_{i=1}^{16} z^2(i)\right] \div 16 \quad .$$

The value of χ^2 is also compared with warning and action limits obtained from tabulated values of the chi-square distribution.

The third comparison computes a reduced chi-square from the m peaks, which have a significantly large net area. First, each A(i) is compared with s(i). If this ratio exceeds 3.0, the peak is included in this analysis. The chi-square from the m peaks, $\chi^2(m)$, is computed

$$\chi^2(m) = \left[\sum_{i=1}^{m} z^2(i)\right] \div m \quad .$$

$\chi^2(m)$ is then compared with both a warning and an action limit. The statistical procedures are taken from Morrison.[2] Beedgen et al.[3] detail the required computations.

VI. RESULTS

The reproducibility obtained from repeated measurements on the same item is consistent with counting statistics. In some cases, the precision from counting statistics is as small as 0.5%. The detection sensitivity of this instrument varies by about 10% along its vertical axis (which is 152 cm high). This variation is expected because four discrete detectors are used. If the sample is moved vertically a small amount, <2 cm, the response varies <2%. A radial offset causes a nonlinear decrease in the measurement response that varies from a 2% decrease at 5 cm offset to a 9% decrease at 10 cm offset. These large effects, caused by the detector collimation, are reduced significantly by rotating the sample during the measurement.

The count rate from the americium seed varies slightly from detector to detector. In addition, there are slight differences in counting efficiency and detector collimation. When these effects are accounted for, the response from four different detectors is independent of detector. Currently, there appears to be a very slight difference in relative efficiency among the four detectors; however, the difference [illegible] too small to be reliably determined from the present data. Table I lists the count rate ratios for a particular gamma-ray energy between a pair of detectors. The five samples vary in enrichment from 17 to 93%. The sigma values are the s(i) estimated from counting statistics. All of the samples had a high-intensity 186-keV signal, but none of the samples had both strong 1001- and 2614-keV signals. Sample number 2 had only a 186-keV signal.

This instrument can sometimes distinguish similar items on the basis of different count rates from the minor isotope (^{232}U, giving rise to the 2614-keV gamma ray) or on the basis of different count rates from the major isotopes (as a result of packaging differences). Otherwise, the items must be sufficiently different to fail one of the comparisons described above.

VII. MEASUREMENT CONTROL

This instrument has several measurement control functions built into it that are intended to help the user determine whether it is functioning correctly. The measurement control functions test for measurement accuracy (bias), precision, and randomness. In addition, the instrument stores the results of a background (no sample) run to be subtracted from each assay. These data should be updated at least daily or whenever the user suspects the room background may have changed. The results of the background run are checked to ensure that the americium seed peak net area is large enough to provide adequate counting precision, that the other peaks are sufficiently small, and that the seed peak has a reasonable centroid and full width at half maximum (FWHM) value. All measurements include the check of at least one peak for an appropriate centroid and FWHM. Most include the check of two peaks.

TABLE I

NORMALIZED COUNT RATE RATIOS
FOR DIFFERENT DETECTORS

		(Normalized to Detector 1)		
Sample ID	Sigma	Det. 2	Det. 3	Det. 4
1	0.020	0.975	1.007	1.090
	0.061	0.995	0.972	0.963
2	0.022	1.109	1.022	0.967
3	0.021	0.979	1.017	1.036
	0.036	0.940	1.018	1.037
4	0.026	1.017	1.031	1.020
	0.067	0.986	0.971	1.109
5	0.023	0.951	0.951	0.910
	0.054	0.924	1.009	1.176

Another measurement control function is a bias check. The user can compare measurements on a known standard and test for a significant difference. A weekly frequency for this check is recommended. We are investigating the possibility of using ^{133}Ba sources to perform this check without using SNM. The precision and reproducibility of a barium source measurement are adequate for measurement control. A long-term evaluation is under way to verify that a change in the detectors' absolute or relative efficiency can be detected in a timely manner with these sources.

Figures 2 and 3 are control charts for the standard deviation of the 356-keV ^{133}Ba peak. The preliminary conclusion is that the scatter might be larger than that predicted from counting statistics. This scatter could be due to sample repositioning errors.

A measurement control function also used in this instrument is a precision check. The user can measure any sample 5 or 15 times. The results for each peak are then individually averaged; a standard deviation is calculated and compared with the predicted standard deviation estimated from counting statistics. The comparison is a chi-square test. A mean-square successive difference (MSSD) test[4] is computed to check for randomness of the data. Individual peak standard deviations are combined to do a chi-square test on the entire set. This measurement control precision test should be done at least monthly to check for unexpected sources of uncertainty or trends in the data.

Table II lists the precision check results obtained over a 30-day period. The longer (1000-s) count time was chosen to evaluate the instruments' behavior under operating conditions more stringent than normal (200-s count time). Both the reduced chi-square, χ^2, and the average MSSD are behaving as expected.

VIII. CONCLUSIONS

The four-gamma confirmatory measurement technique will provide a cost-effective alternative to additional physical security measures in the problem of S/R differences. It will also help separate the issues of material control and measurement control by the timely measurement of an attribute unique to the SNM of interest. We hope that this will eventually lead to increased attention in the area in which most of us believe S/R differences occur: measurement control.

There are several statistical tests for the measurement control functions inherent in this instrument and for the S/R comparison. The use of so many tests can result in an increased false alarm rate, which requires time and expense to resolve. Thus, it is clear that the false alarm rate for this instrument must

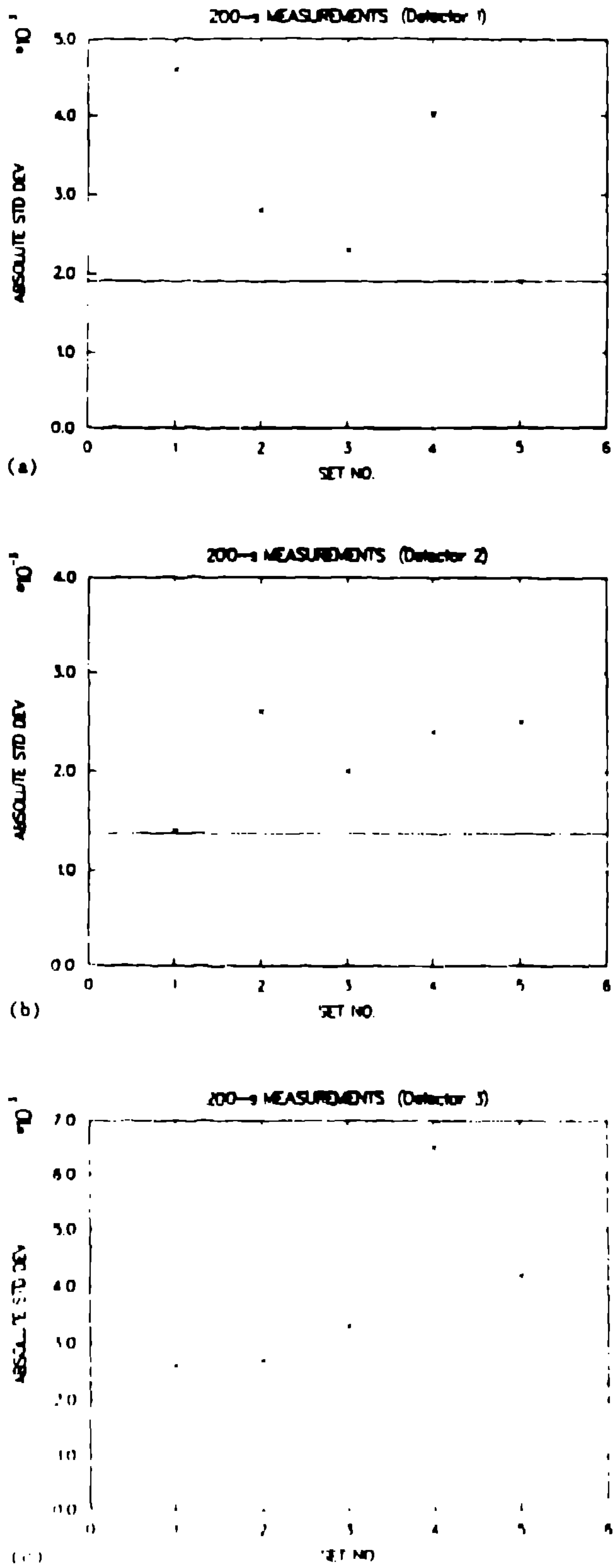

Fig. 2(a)-(c). Control charts for the standard deviation of the background-corrected net peak area of the 356-keV ^{133}Ba peak during 200-s runs for each of three detectors. Each point is the standard deviation of five independent counts. The solid line is the expected value based on counting statistics; the dashed line is the 3σ control limit.

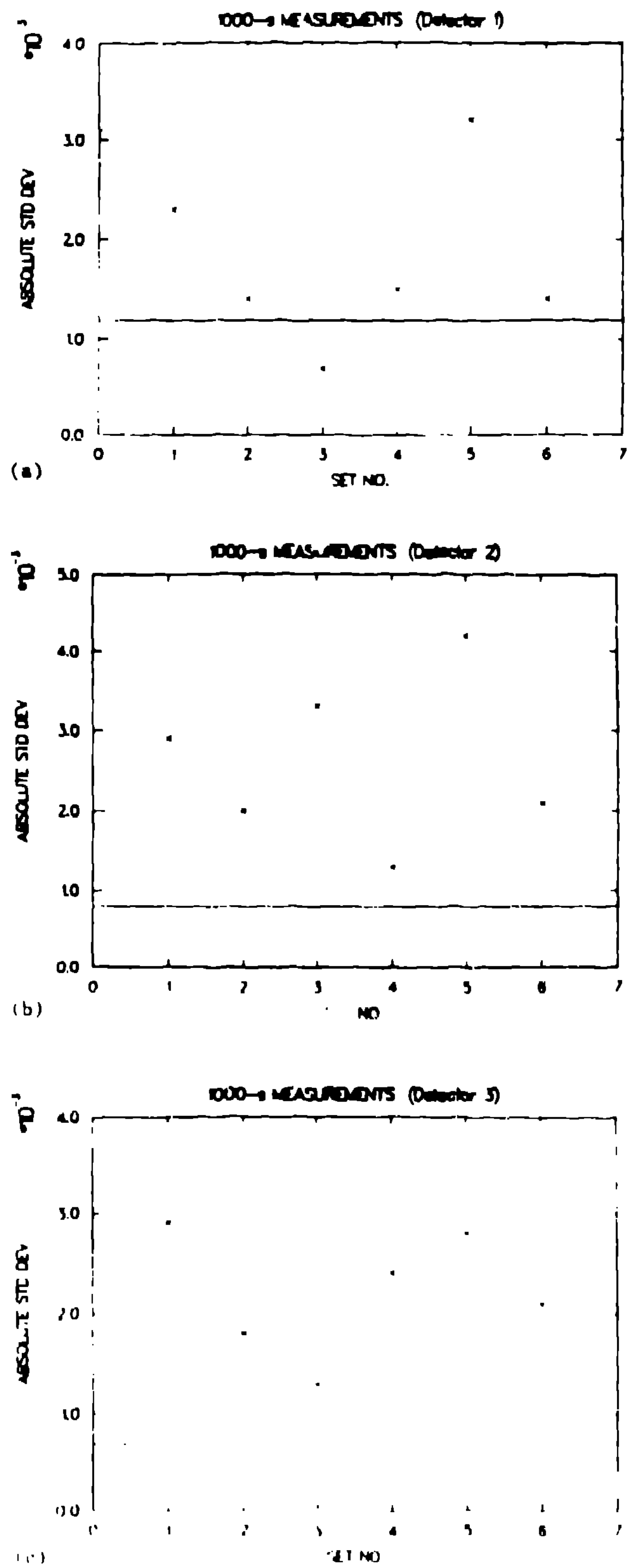

Fig. 3(a)-(c). Control charts for the standard deviation of the background-corrected net peak area of the 356-keV ^{133}Ba peak during 1000-s runs for each of three detectors. Each point is the average of three independent counts. The solid line is the value predicted from counting statistics; the dashed line is the 3σ control limit.

TABLE II

PRECISION CHECK RESULTS

χ^2	MSSD[a]	Count Time (s)	Number Runs
1.18	0.79	1000	5
0.95	-0.35	1000	5
0.83	0.91	1000	5
1.12	1.093	1000	5
1.02	0.27	1000	5
1.17	-0.23	1000	5
1.28	0.99	1000	5
0.98	2.68	200	5
1.19	1.23	200	5
1.01	-0.08	200	15
1.11	-0.12	200	15
0.90	0.92	200	5

[a]MSSD = Average over 16 peaks of mean-square successive difference.

be very low if it is to be useful to plant operators. A few false alarms during a year will rapidly cause most of the plant operators to respond as if they expect a false alarm rather than a true diversion. This instrument cannot do much to correct mislabeled results or results that are interchanged between two samples. (If this trend is currently a problem, this instrumentation should direct attention to solving this problem in a timely fashion.) The action limits, however, can be chosen (that is, raised) to minimize the false-alarm rate.

As an illustration of the possibilities of a large false-alarm rate, consider n independent tests with a 5% false-alarm probability. The overall false-alarm probability for n = 5 tests is 22%, for n = 10 tests is 40%, and for n = 20 is 64%. Thus, the false-alarm probability can increase quickly for multiple independent tests. Consequently, the action limits for each test must be kept very conservative.

However, the conservative action limit does not limit the effectiveness of this technique as much as a cursory overview might suggest. If the four-gamma technique is considered in the context of the rest of the system's safeguards, it will be used to detect large diversions in a rapid manner. If an individual penetrates the system of barriers, seals, and other protective measures, that person will probably tamper with a small number of items in some gross fashion (for example, take the entire item). That type of tampering does not require a high precision, highly accurate measurement for detection. If a trickle diversion scenario is chosen, the length of time the system is penetrated is much greater. The greater length of time required for a successful diversion will be more susceptible to detection by other means, such as input accountability measurements or physical security.

In addition, we must remember that the precision of this measurement is much better than its accuracy. We are accustomed to thinking about a comparison between two results in terms of the accuracies of the two different types of measurement. In the case presented here, the measurement precision can often allow the user to distinguish between two similar items in terms of counts per second, while an attempt to calibrate in grams brings in other sources of measurement uncertainty such that the user can only determine that the two items are similar. In fact, two measurements of the same generic type are susceptible to identical calibration errors, which cancel each other in the comparison proposed for this type of measurement.

IX. FUTURE PLANS

The present plans for this instrument are to install it at the Y-12 Plant at Oak Ridge National Laboratory for an evaluation during 1988. The instrument will reside in a dedicated counting room surrounded by 60-cm-thick concrete walls. The massive shielding is required because the counting room is inside the receiver area, which is physically contained in, but separated from, the storage vault. The receiver area has the capacity to store approximately 100 shipping containers awaiting measurement. During the evaluation at Y-12, we plan to investigate the effects of handling the barrels, of interchanging detectors, and of packaging. If the evaluation is successful, a second instrument is planned for installation at a designated shipper facility. Two instruments at two facilities will be needed to evaluate the actual measurement scenario, complete with practical problems such as calibration and settling of the container contents during shipment.

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