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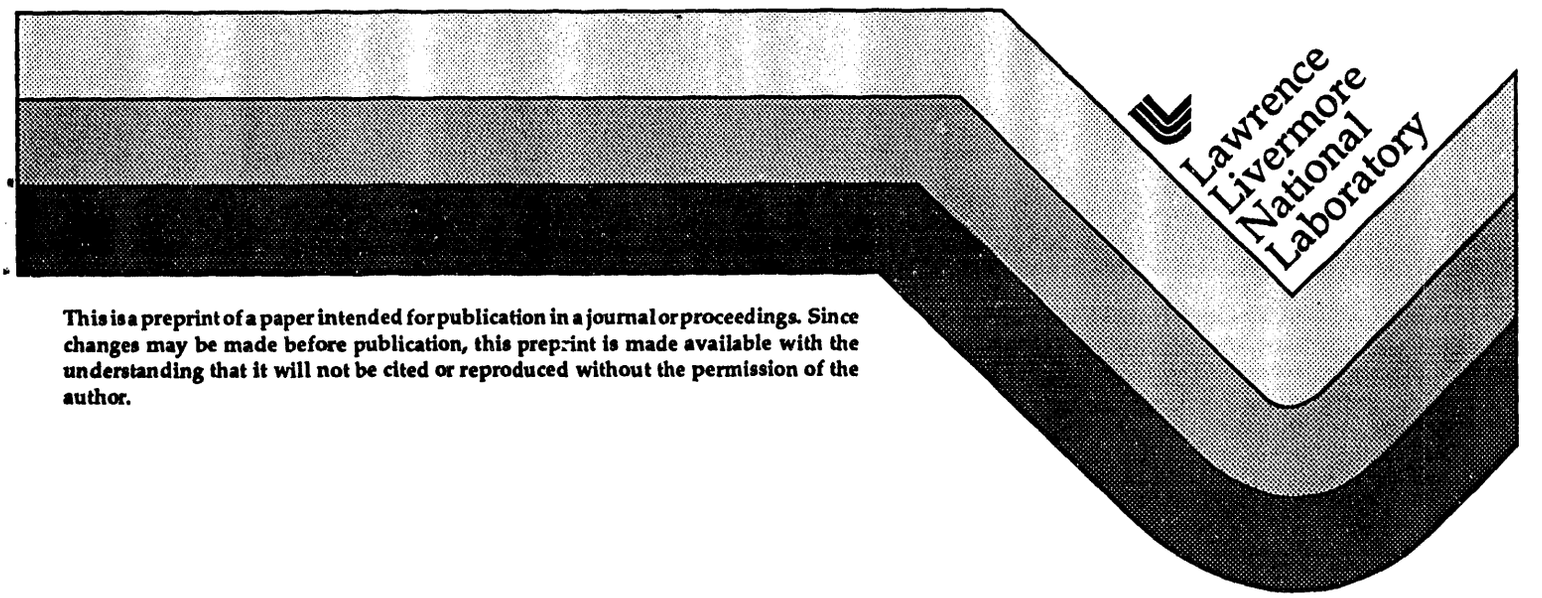

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Application of Cadmium-Zinc-Telluride Detectors in U-235 Enrichment Measurements*

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Abstract

High-resolution, gamma- and x-ray spectrometry are used routinely in nuclear safeguards verification measurements of plutonium and uranium in the field. These measurements are mostly performed with high-purity germanium (HPGe) detectors, that require cooling to liquid-nitrogen temperatures, thus limiting their utility in field and unattended safeguards measurement applications. NaI scintillation detectors do not require cooling, but their moderate energy resolution (10% at 122 keV) is insufficient in most cases for reliable verification measurements. Semiconductor detectors that operate at room temperature, such as cadmium-zinc-telluride (CdZnTe) detectors, with energy resolution performance reaching 2.0% at 122 keV may complement HPGe detectors for certain safeguards verification applications. We used a 5x5x5 mm CdZnTe detector to measure U-235 enrichments ranging from 3% to 75%. We use a spectrum analysis technique that fits U-235, U-238, and U K x-ray response profiles to data in the 89- to 100-keV region of a gamma-ray spectrum. From the relative magnitudes of the U-235 and U-238 profiles we determine the U-235 enrichment with an accuracy of about 10% with CdZnTe detectors.

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1. Introduction

The traditional method used for ^{235}U enrichment measurements, developed more than 20 years ago [1], is based on measurement of the 186-keV peak of ^{235}U. This method utilizes the "enrichment-meter" principle where the fundamental relationship is that the intensity of the 186-keV gamma rays emitted from a fixed area of a thick sample is proportional to the enrichment of the sample. "Thick" in this context means several mean free paths at 186 keV. Although the measurement technique is simple in concept and can be applied with NaI-based systems for field-portable applications, it requires calibration and also that unknown items meet the calibration conditions.

Recently, there has been a need for determining ^{235}U enrichments in items where no suitable reference standards exist or where non-reproducible conditions make calibration impossible. Gunnink [2] has developed a gamma-ray analysis technique, incorporated in a computer program called MGAU, to determine ^{235}U enrichments by analyzing the gamma- and x-ray peaks in the 88 to 100-keV region of a uranium spectrum. An advantage in analyzing this energy region is that the ^{235}U and ^{238}U photon emissions are very close in energy and therefore detected with comparable efficiencies. This analysis approach eliminates the need for calibration and allows ^{235}U enrichment measurements in arbitrary samples.

This region of the uranium spectrum has a complex structure with closely overlapping peaks which is the greatest obstacle to accurate analyses. To achieve high precision and accuracy, this analysis requires good energy resolution as can be seen in Figure 1 in which the spectral data were measured with a small, planar high-purity (HPGe) germanium detector

with energy resolution of .5% at 122 keV. With this analysis technique, accuracies better than 1% have been obtained with ^{235}U enrichments ranging from natural to 93%.

HPGe detectors have limited utility in field and unattended safeguards measurement applications, because they require cooling to liquid nitrogen temperatures. NaI detectors do not require cooling, but their moderate energy resolution is insufficient for application of Gunnink's new uranium spectrum analysis technique. We have begun to explore the use of semiconductor detectors that operate at room temperature, such as cadmium-zinc-telluride (CdZnTe) detectors, with energy resolution performance reaching 2.0% at 122 keV. Such detectors may complement HPGe detectors for certain safeguards measurements where portable or unattended measurement equipment is needed.

We have applied an analysis technique similar to that developed for HPGe detectors to the 100-keV region of uranium spectral data measured with a CdZnTe detector. Figure 2 shows an analysis of the peak grouping in the 88- to 100-keV region for the same ^{235}U enrichment of 10% as in Figure 1. We have measured uranium standards with 3, 5, 10, 50, and 75 per cent ^{235}U enrichment using a CdZnTe detector. In this report we present the results of these measurements.

2. Summary of Method

The general method for determining relative isotopic abundances from gamma-ray spectra is to measure and interpret gamma- and x-ray peak intensities associated with the different isotopes. Relative isotopic abundances can be calculated with known branching intensities and half-lives for the isotopes and applying corrections for the differences in detection efficiencies of the measured peaks. The relative efficiency factors can usually be determined

from the measured spectrum, if the peaks are close in energy.

When the peaks severely overlap each other, the determination of the peak intensities becomes more complicated; then, peak-fitting techniques must be used. Sometimes the overlap is so severe and the energy resolution is such that special steps must be taken in the fitting process to obtain proper convergence. In such circumstances, not all of the peak positions and heights can be allowed to be independently free in the fitting process. To overcome these difficulties, we have developed a "response function" method that allows peaks belonging to a given isotopic component to have their energies and heights to be fixed relative to each other. We also fix other peak parameters like those which describe the low-energy exponential tail used in our model of a gamma- and x-ray peaks. These parameters can be predetermined from other isolated peaks in the spectrum. This model and the fitting methods described here are incorporated into a computer program called GRPANL developed by the authors.

We have applied these techniques to the peaks in the 89- to 100-keV region of the uranium spectrum. The 89- and 93-keV Th x-ray peaks from the decay of ^{235}U and the 92.367- and 92.792-keV gamma-ray peaks from ^{238}U-^{234}Th are very close in energy and are ideally suited for determining the relative abundances of ^{235}U and ^{238}U. The 94- and 98-keV uranium x-ray peaks are due primarily to self-induced fluorescence of the uranium sample. Note that the gamma- and x-ray peak shapes are different and require different peak-shape algorithms, which are incorporated into the GRPANL program. In the MGAU analysis of uranium spectra taken with a germanium detector, corrections are made for the small differences in detection efficiencies between the various peaks in this region. This is done

by comparing the measured intensities of the K_α and K_β x-ray peaks of uranium, which are always present due to self-induced fluorescence of the sample. We did not correct peak intensities from spectra taken with CdZnTe detectors for differences in relative detection efficiencies, because of the difficulties in analyzing the K_β region.

In the 87- to 101-keV region of the spectrum, we fit peaks with the following energies to the net channel counts: 89.96, 90.0, 92.27, 92.376, 92.792, 93.36, 94.66, 95.89, 98.443, and 99.27 keV. We fix the positions of these peaks relative to the position of the U $K_{\alpha 1}$ x-ray peak in the fitting process. We also fix the intensity of those peaks from the ^{235}U decay relative to the 93.3-keV Th $K_{\alpha 1}$ x-ray peak intensity; we fix the intensity of the ^{234}Th 92.792-keV gamma-ray peak relative to the ^{234}Th 92.376-keV gamma-ray peak; and the U $K_{\alpha 2}$ x-ray intensity is fixed relative to the U $K_{\alpha 1}$ x-ray intensity. In the fitting process we allow the following five parameters to be free: 1) the 98.443-keV peak position and 2) height, 3) the 92.376-keV peak height, 4) the 93.36-keV peak height, and 5) the Gaussian full-width-at-half-maximum (FWHM). The two parameters which describe the gamma-ray peak shape, 1) the low-energy exponential tail slope and 2) amplitude, are determined from the 185.7-keV gamma-ray peak in the spectrum and are fixed in the fitting process.

3. Results

The analysis methods summarized here are incorporated into an analysis control file used by the GRPANL program. To evaluate the applicability and accuracy of this analysis method on uranium spectrum acquired with CdZnTe detectors, we acquired spectra of various uranium standards. One-gram uranium oxide, U_3O_8, reference materials from NBL and isotopic standards from NBS with ^{235}U atom abundances of 3.05, 5.01, 10.19, 49.7, and 75.36

per cent were measured. A 5x5x5 mm CdZnTe detector operated at 1000 volts bias was used for our measurements. We used the detector with an Ortec 142PC preamplifier and a Canberra 2020 amplifier with a Gaussian shaping time of 0.5 microseconds. The spectra were measured with a computer-based multichannel analyzer. Spectra were taken with a gain of 75 eV per channel. An energy resolution (FWHM) of 2.4 keV at 122 keV was obtained.

We placed samples on the face of the detector and counted them for 6, 12 or 15 hours. At ^{235}U enrichments of 3%, 50%, and 75% it was necessary to count for the longer times to obtain sufficient counts in the peaks of the lesser of the two major uranium isotopes (^{235}U and ^{238}U). A 6-hour measurement of the one-gram uranium sample with an ^{235}U enrichment of 10% gave $3.4x10^6$ total counts in an energy spectrum from 0 to 300-keV. The ^{234}Th 92.8-keV gamma-ray and Th 93.3-keV x-ray peaks in this spectrum had 31,000 and 46,000 net counts, respectively. Table 1 compares the measured ^{235}U to ^{238}U ratio to the declared ratio for the five samples measured with the 5x5x5 mm CdZnTe detector.

4. Conclusions

The results shown in Table 1 indicate that ^{235}U enrichments can be determined with the described data-analysis approach on gamma-ray spectra with energy resolutions of 2.0% at 122 keV. Accuracies on the order of 10% can be obtained on ^{235}U enrichments from 3% to 50% using CdZnTe detectors with our data-analysis methods. Our measurements also show that larger CdZnTe detectors are needed to reduce measurement times, which are one hour or less with a 200 mm^2, planar HPGe detector for the samples used in this study

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References

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[2] R. Gunnink, W.D. Ruhter, P. Miller, et. al., "MGAU: A New Analysis Code for Measuring U-235 Enrichments in Arbitrary Samples," Lawrence Livermore National Laboratory, UCRL-JC-114713 (1994).

[3] R. Gunnink, W.D. Ruhter, and J.B. Niday, "GRPANL: A Suite of Computer Programs for Analyzing Complex Ge and Alpha-Particle Spectra," Lawrence Livermore National Laboratory, UCRL-53861, Vol.1 (1988).

Figures

Fig. 1 This plot of the 88- to 100-keV region of a uranium spectrum with a ^{235}U enrichment of 10% obtained with a high-resolution germanium detector shows the component responses for ^{235}U, ^{238}U, and U x rays. Each response is calculated from known nuclear parameters and from energy scaling and peak width information determined from other gamma-ray peaks in the spectrum. ^{235}U enrichment is determined from the relative amounts of the ^{235}U and ^{238}U responses.

Fig. 2 This plot shows an analysis similar to that shown in Fig. 1 except the spectral data were obtained with a 5x5x5 mm CdZnTe detector with an energy resolution of 2.4 keV at 122 keV.

Declared ^{235}U / ^{238}U	Measured ^{235}U / ^{238}U $\pm$ %RSD	Measurement time (hours)
.0314	.0302 $\pm$ 16.8	12
.0528	.0557 $\pm$ 12.5	6
.1136	.1116 $\pm$ 4.6	6
.9997	1.0093 $\pm$ 13.7	15
3.166	4.42 $\pm$ 36.3	15

Table 1. Comparison of measured ^{235}U to ^{238}U ratios with declared ratios. Relative standard deviations (RSD) were determined from the fits to the net counts in the spectra.

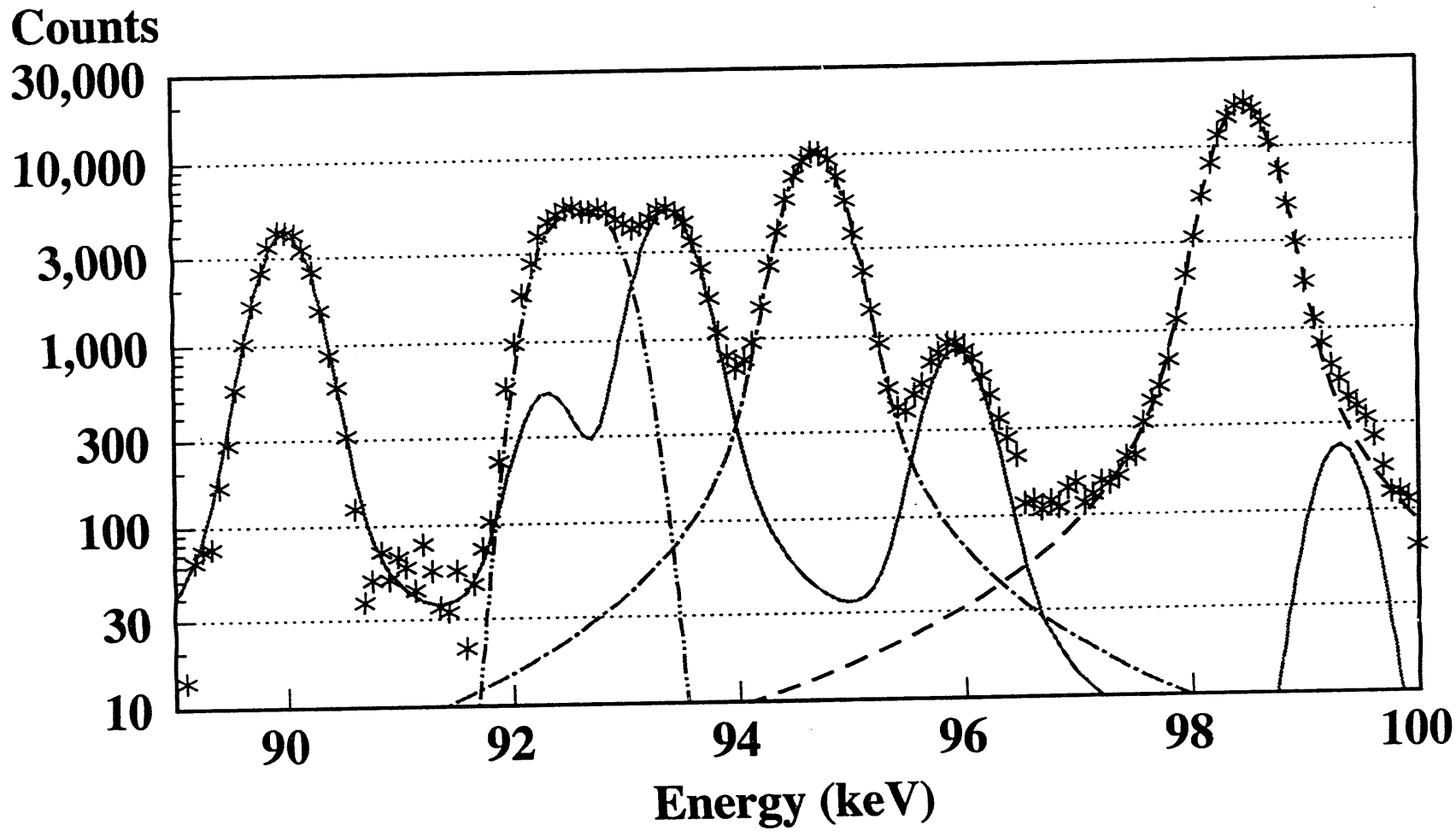
Counts
30,000
10,000
3,000
1,000
300
100
30
10
90
92
94
96
98
100
Energy (keV)
Net Counts
U-235
U-238
U X-ray
U X-ray

Figure 1

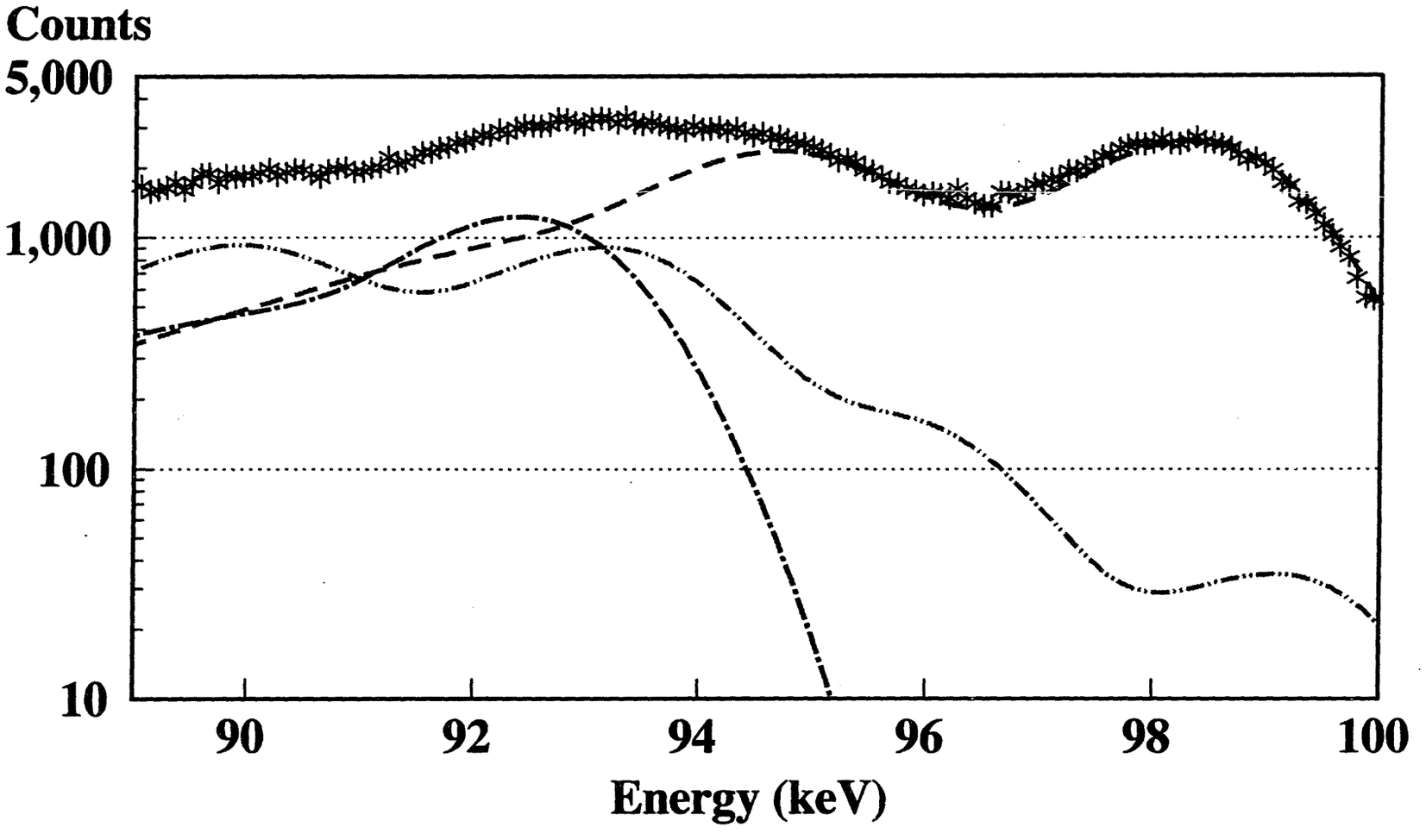
Counts
5,000
1,000
100
10
90
92
94
96
98
100
Energy (keV)
Net Counts
U-235
U-238
U X-ray

Figure 2

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