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CORRECTIONS FOR THE EMISSIONS PROBABILITIES OF THE INTENSE GAMMA RAYS FROM URANIUM-238 AND APPLYING THEM TO URANIUM ISOTOPIC ANALYSIS

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Corrections for the Emission Probabilities of the Intense Gamma Rays from ^{238}U and Applying them to Uranium Isotopic Analysis

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Abstract

Gamma-ray emission probabilities of ^{235}U and ^{238}U have been studied extensively. While the gamma-ray emission probabilities for ^{235}U are well known, the emission probabilities for ^{238}U differ widely from one study to another. Our study indicates that one of the causes of the large discrepancies in the studies may be due to the true coincidence summing of many gamma rays from the decay of ^{238}U daughters. The intensities of some of the intense gamma rays of ^{238}U compared with the others were reduced by as much as 16% when the 21-mm-thick U_3O_8 samples were moved from 15 cm to putting them in contact with the detector. With better knowledge of the gamma-ray emission probabilities and understanding the summing effects of specific gamma rays, the isotopic concentration of uranium can be better analyzed. The results of the isotopic analysis using fixed energy response function analysis with multiple efficiencies (FRAM) with the summing correction are presented in this paper.

I. INTRODUCTION

Material control and accountability (MC&A) is an important part of nonproliferation and safeguards. It requires the precise knowledge of nuclear measurement, not only for data evaluation but also for project management and auditing strategies. As a result of increased fissile MC&A operations in the U.S. and around the world due to the post-Cold War reduction in weapons manufacturing and the increase in weapons dismantlement, new techniques of isotopic measurements are needed and are being developed.

In the past several years, nondestructive assay (NDA) techniques have been developed for uranium enrichment measurements. Most of these NDA methods involve measuring x-rays and gamma rays, which require the precise knowledge of the gamma-ray emission probabilities, particularly those of ^{235}U and ^{238}U. The branching ratios (BRs) of ^{235}U are well known. However, the BRs of ^{238}U and its daughters vary widely in their values and precision. Uranium-238 decays to ^{234}Th by emitting an alpha particle. Thorium-234, with a half-life of 24 days, in turn decays to ^{234}Pa by emitting an electron, and the excited ^{234}Pa, with the half-life of 1.2 minutes, decays to ^{234}U also by emitting an electron. All of the gamma rays of ^{238}U daughters discussed in this paper come from the ^{234}Pa decay to ^{234}U. The most accurate (in the authors' opinion) study of the emission probabilities of ^{238}U and its daughters was that of Scott and Marlow [1], and this study was very different from the values in the latest Table of Isotopes [2] which includes the results of Ref. [1] in the compilation. The BRs of the 766- and 743-keV gamma rays (second and third strongest peaks) from the daughter ^{234m}Pa decay from Ref. [1] are about 10% greater than those in Ref. [2], well outside the limits of their uncertainties. The cause of the large discrepancies between the studies is believed to come from the true coincidence summing of many gamma rays in ^{238}U.

The purpose of this study is to precisely determine the emission probabilities and the intensity-attenuation due to the summing of the most intense gamma rays from the ^{238}U decay chain (and also that of ^{235}U). This would allow us to analyze the uranium isotopes more accurately.

II. APPARATUS

A. Sources

Two certified U_3O_8 samples in cylindrical aluminum cans (7-cm diameter x 2.1-cm depth) with a ^{235}U mass abundance of 0.7119 $\pm$3% and 4.4623 $\pm$16% were used in the experiment (NBS-SRM969/EC-NRM171) [3]. Each can contains 200 g of U_3O_8 powder. The 4.4-g/cm^2 uranium surface density of the samples can be considered infinitely thick for the gamma rays with energies of several hundred keV.

B. Detector and Electronics

The detector used in the experiment was a 2-year-old HPGe coaxial detector manufactured by Canberra with a relative efficiency of about 25%. The detector was shielded with lead bricks and cadmium sheets (except the front) to reduce the background gamma rays. The electronics system was a standard nuclear instrumental methods (NIM) system which consisted of an Ortec 4002D power supply, a Canberra 3106D HV power supply, an Ortec 672 Spectroscopy Amplifier, a Canberra 8077 Fast ADC, a Canberra 8232 Digital Stabilizer, and a Canberra 2071AS Dual Counter Timer. The Canberra 4610 System 100 MCA board was used for the interfacing. The IBM Docking Station I was used to hold the MCA board, and the host computer was an IBM ThinkPad 760L. The software used for collecting data was the System 100 software from Canberra in combination with Fixed energy Response function Analysis with Multiple efficiencies (FRAM) isotopic analysis code from Los Alamos National Laboratory [4][5][6].

III. DATA ACQUISITION

Two sets of data were collected for each of the two sources. One set was with 1.2 mm of cadmium plus 0.4-mm copper absorbers placed in front of the detector, and the other set was without any absorber. The data were obtained with the sources placed at 0, 1, 3, 5, 10, and 15 cm from the detector's front edge. (As for the data sets with the absorbers, the effective distance from the front edge of the sources to the front edge of the detector would be about 2 mm when the sources are pressed against the absorbers.) For the 0.71% ^{235}U data collection, the shaping time of the amplifier was set at 6 μs with a triangular shape to achieve better energy resolution. For the 4.5% ^{235}U, the count rates were much higher due to the abundance of ^{235}U gamma rays, and the shaping time was reduced to 2 μs. The 186-keV peak of ^{235}U and 1001-keV peak of ^{238}U were used for stabilization. Twenty-one 1-h spectra were taken at each configuration, and they were summed into one single spectrum to simplify the analysis.

IV. DATA ANALYSIS

A. Results Using FRAM with Current Parameter File

The data were first analyzed using FRAM, a code developed at Los Alamos for isotopic analysis based on a self-calibration using several gamma-ray peaks for determining the isotopic ratios. For low-enriched uranium isotopic determination, the current parameter uses the 144-, 163-, 186-,

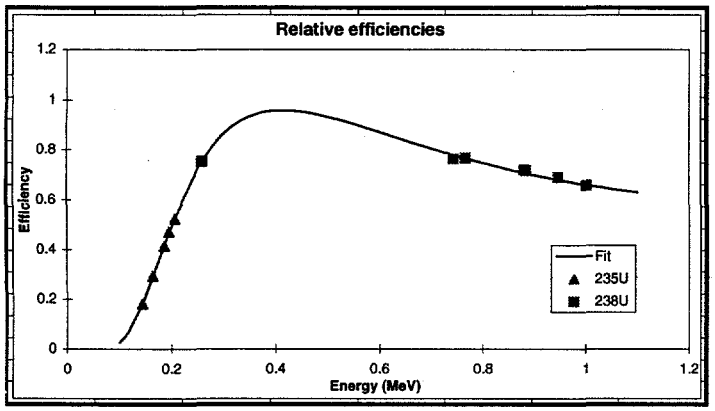

Fig. 1. Example of a relative efficiency curve of low-enriched uranium.

195-, and 205-keV peaks for ^{235}U and 258-, 743-, 880-, 883-, 946-, and 1001-keV peaks for ^{238}U and its daughters to determine the relative efficiencies. Fig. 1 shows a typical relative efficiency curve of low-enriched uranium. From the peaks used, there is no overlap of peaks between the gamma rays of ^{235}U and ^{238}U. Also, a large gap exists between the 258- and 742-keV peak of ^{238}U. This, in effect, makes the 258-keV peak the driving force for fitting the relative efficiency curve, and therefore, it is the only peak used to identify the activity of ^{238}U. Because the 258-keV peak is the only peak used in determining the activity of ^{238}U, the $^{235}U/^{238}U$ ratio depends mostly on determining the 258-keV peak. If the BR of the 258-keV peak is not correct or if some of its area underneath its peak in the spectrum is lost due to summing, then the $^{235}U/^{238}U$ ratio will not likely be correct.

The results of the analysis are shown in Fig. 2. Clearly, the $^{235}U/^{238}U$ mass ratio is a function of the distance from the detector. This dependence is because the 258-keV peak loses some counts due to the summing effect. The 258-keV gamma ray is strongly in coincidence with many other peaks. When it enters the detector together with one of its coincident gamma rays, the resulting pulse from the detector will not correspond to 258 keV but to the sum of the two energies or their Compton energies. The result is the loss of counts underneath the 258-keV peak. As the source is moved farther away from the detector, the solid angle of the detector as seen by the source is reduced and so are the chances for two gamma rays to enter the detector at the same time. Therefore, the $^{235}U/^{238}U$ mass ratio curve is flattened out as the distance between the source and the detector increases.

B. Correction for the Summing Effect

The certified values of the $^{235}U/^{238}U$ activity ratios were calculated using the equation

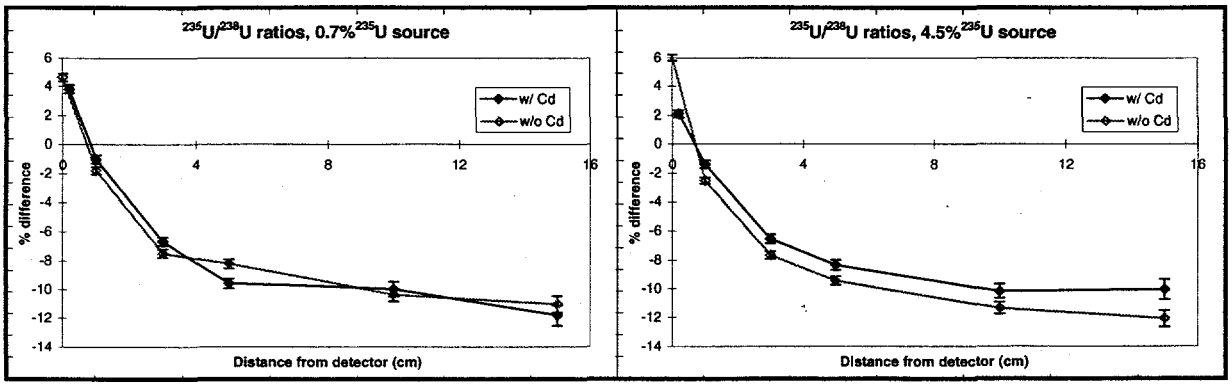

Fig. 2. Results of FRAM using the current parameter file. The vertical scales show the differences between the calculated ratios and the accepted ones where difference = measured/accepted -1.

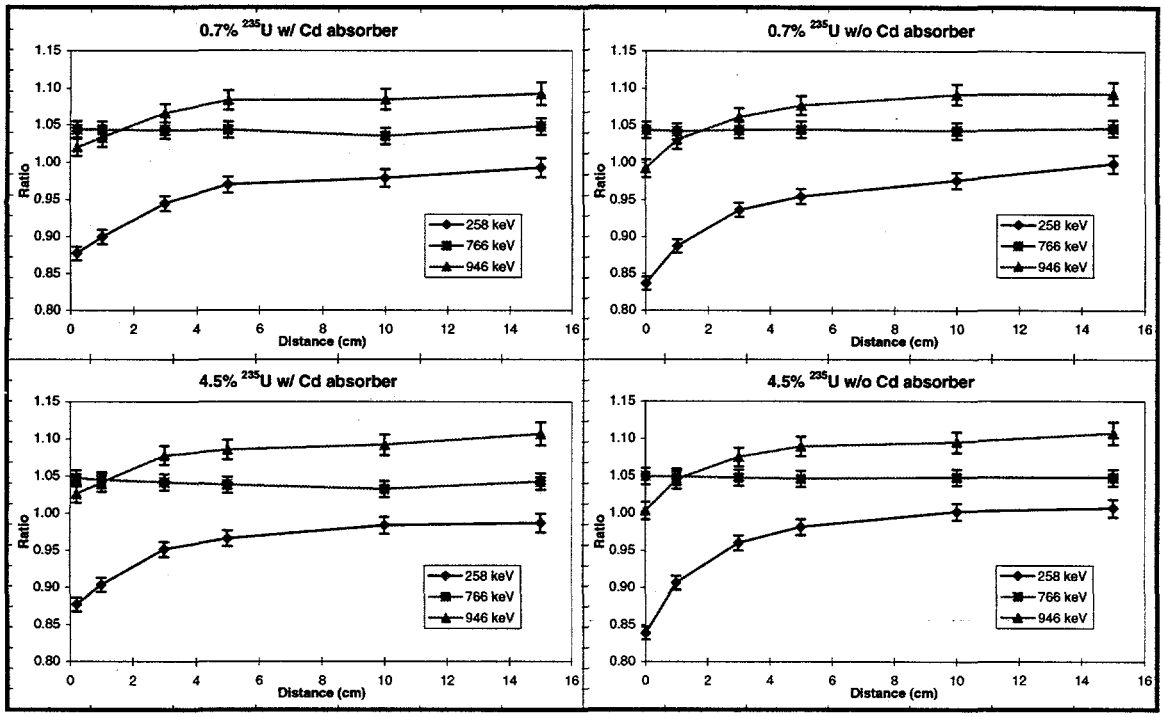

Fig. 3. The ratios of some of the measured peak areas/BRs of ^{238}U to those from the fits. The curvature of the lines represent the fractional intensity-attenuations of the peaks. The ratio values of the curves at infinity represent the ratios of the correct BRs to the BRs used in the areas/BRs calculations.

$$\text{Activity ratio} = \text{Mass ratio}/[(\text{Half-life ratio}) \times (\text{Atomic mass ratio})].$$

For the sources used, the activity ratios turned out to be 0.04610 and 0.30046 for the 0.7% and 4.5% ^{235}U sources respectively.

The peak areas determined from FRAM are used as the first step to correct for the summing effect. The peak areas of ^{235}U are normalized to those of ^{238}U by dividing by the activity ratios. The efficiency curve is obtained by fitting some of the normalized peak areas to the equation

$$\text{Area/BR} = (\text{Activ}) \times (\text{Self atten}) \times (\text{Cd+Cu atten}) \times (\text{Det eff}) \times (\text{Correction factor}).$$

Rearrange the equation, then

$$(\text{Area/BR})/[(\text{Self atten}) \times (\text{Cd+Cu atten}) \times (\text{Det eff})] = (\text{Activ}) \times (\text{Correction factor}).$$

The activity multiplied by the correction factor is set equal to $aE^bc^{1/E}$, where E is the energy of the peak in units of MeV; a, b, and c are constants.

The correction factor $aE^bc^{1/E}$ was found to work well in correcting for the deviations of the self-attenuation, absorber attenuation, and of the detector efficiency. The 144-, 163-, 186-, and 205-keV peaks of ^{235}U and the 1001-keV peak of ^{238}U were used in the fit. The BRs of these peaks were obtained from Ref. [2]. The attenuation cross sections of uranium and oxygen in U_3O_8 were obtained from Ref. [7]. The attenuation of the cadmium and copper absorbers were measured directly. The detector efficiency was also obtained using the ^{133}Ba and ^{152}Eu sources.

The variance-covariance matrix from the fit was used to estimate the uncertainties of the relative efficiency curve predicted by the model. The ratios of the measured peak areas/BRs to those from

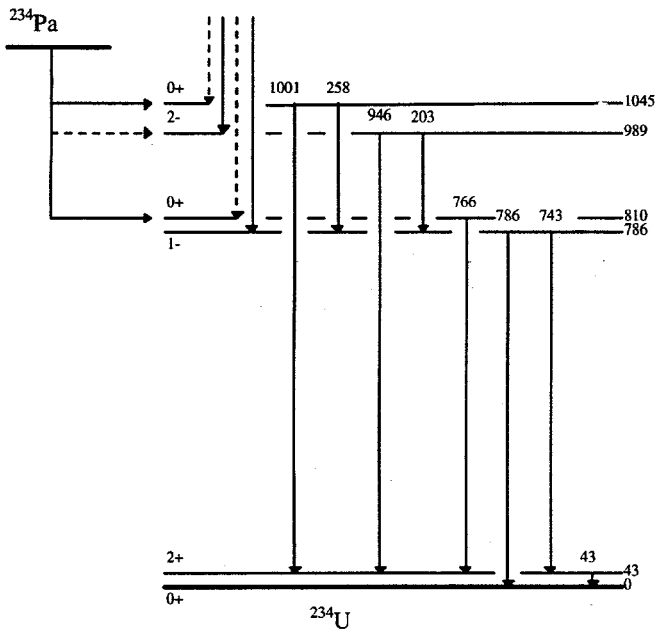

Fig. 4. Partial decay scheme of the ^{234}Pa daughter of ^{238}U. The dotted lines show the weak feeding transitions. The levels are not drawn to scale.

the fit were calculated. Fig. 3 shows the ratios for some of the intense peaks of ^{238}U. The 258- and 946-keV peaks lose a large fraction of their intensity, up to 16%, when the source is moved close to the detector while for the 766-keV peak, its intensity remains constant regardless of the source-detector distance. Along with the intensity-attenuation, one can see that the curves resemble the exponential function and are flattening out as the source is moved farther away from the detector. At infinity, the curves would be flat and their values would represent the unattenuated intensities of the peaks. These values should also be equal to one if the BRs used in the areas/BRs calculations are correct. Any deviation from one would mean that the BR is incorrect and should be corrected.

The reason the 258- and 946-keV peaks lose intensity while the 766-keV does not lose intensity can be explained with the help of Fig. 4. Figure 4 shows the partial decay scheme of some of gamma rays of the ^{234}Pa daughter of ^{238}U. The 766- and 1001-keV transitions decay from levels which are fed mostly from the direct decay of ^{234}Pa. Therefore, most of the 766- and 1001-keV gamma rays are not in coincidence with any other gamma rays except the 43-keV peak. Because of the very low energy, most of the 43-keV gamma rays are absorbed by the surrounding absorbers and the material itself, and only a very small fraction can get to the detector to be in coincidence with the 766- or 1001-keV gamma ray. This explains why the 766-keV peak does not lose intensity due to summing as seen in Fig. 3. For the 258-keV peak, it is in coincidence with the 743- and 786-keV and gamma rays. If the 258-keV gamma ray enters the detector together with one or more gamma rays, then the output pulse from the detector will be that of the sum energy (or the Compton of the sum energy) and not that of 258 keV. As for the 946-keV transition, it decays from the 989-keV level which is fed in part by the direct decay of ^{234}Pa and in part by the levels above it. This results in the 946-keV transition being in coincidence with many other gamma rays. Thus, it is also affected by the summing effect. The argument for all other gamma rays from ^{234}Pa decay can be similarly explained. As for the reason the 810- and 1045-keV levels (the levels in which the 766- and 1001-keV transitions originate) receive a minute amount of feeding from the levels above it while the other levels receive much more can be explained as follows. These two levels have the spins of 0+, while all other levels (except the ground state) have non-zero spin. Most of the levels above them have large spins (three or more). To decay to the zero-spin states requires either E3 or M3 or higher multipole transition, and this probability is much smaller than to decay to non-zero states which may require either E2 or M2 or lower multipole transitions.

Table I. The branching ratios and relative fractional intensity-attenuations of some of the gamma rays used in uranium isotopic determination.

Isotope	Energy	BR w/ Cd	err w/ Cd	BR w/o Cd	err w/o Cd	*b/a* w/ Cd	err w/ Cd	*b/a* w/o Cd	err w/o Cd
^{234}U	120.90	3.548E-04	4.9E-06	3.433E-04	4.2E-06	0.000	0.021	0.012	0.016
^{235}U	143.76	1.095E-01	3.4E-04	1.095E-01	3.1E-04	0.000	0.006	0.002	0.005
^{235}U	163.33	5.087E-02	1.1E-04	5.090E-02	1.0E-04	0.001	0.004	0.000	0.004
^{235}U	185.72	5.734E-01	1.3E-03	5.740E-01	1.4E-03	0.000	0.005	0.004	0.004
^{235}U	194.94	6.292E-03	2.6E-05	6.247E-03	2.2E-05	0.002	0.007	0.034	0.016
^{235}U	202.11	1.095E-02	4.2E-05	1.094E-02	6.3E-05	0.035	0.008	0.131	0.015
^{235}U	205.31	4.994E-02	1.6E-04	4.992E-02	1.5E-04	0.000	0.006	0.000	0.006
^{238}U	258.26	7.191E-04	4.0E-06	7.259E-04	6.3E-06	0.122	0.009	0.156	0.009
^{238}U	568.80	5.713E-05	4.1E-07	5.762E-05	3.9E-07	0.087	0.011	0.131	0.012
^{238}U	569.40	1.305E-04	9.4E-07	1.316E-04	8.9E-07	0.087	0.012	0.131	0.012
^{238}U	742.81	8.913E-04	4.1E-06	8.969E-04	4.6E-06	0.035	0.010	0.056	0.008
^{238}U	766.36	3.061E-03	1.4E-05	3.076E-03	1.4E-05	0.000	0.009	0.001	0.008
^{238}U	786.27	5.393E-04	2.6E-06	5.403E-04	2.5E-06	0.049	0.009	0.068	0.008
^{238}U	880.45	2.092E-04	1.3E-06	2.114E-04	1.8E-06	0.060	0.011	0.084	0.016
^{238}U	883.22	2.114E-04	1.9E-06	2.102E-04	1.7E-06	0.074	0.015	0.091	0.012
^{238}U	945.96	3.434E-04	2.2E-06	3.442E-04	1.9E-06	0.073	0.010	0.089	0.009
^{238}U	1001.03	8.371E-03	4.2E-05	8.371E-03	4.1E-05	0.000	0.010	0.000	0.009

To obtain the new BRs and the relative intensity-attenuations, one can just fit the individual measured-to-fit ratios of each energy peak to the exponential equation [$y = a\text{-}be^{-cx}$], where x represents the distance from detector, y represents the ratio of the measured values to those of the fit found earlier, and *a*, *b*, and *c* are some unknown, positive variables. At infinity, $y = a$ which would correspond to the ratio of the corrected BR to the old BR. The value *b/a* represents the fractional intensity-attenuation at distance of zero. The intensity-attenuation coefficient *c* is a function of the geometry of the detector and of the source and should be the same for all the energy peaks.

When fitting the individual data of each energy peak to the above equation, especially those with small intensity-attenuations, the intensity-attenuation coefficient *c* varies from energy set to energy set. It is, therefore, better if a common intensity-attenuation coefficient *c* is found and used for all the individual fits.

The data of nine intense peaks of ^{238}U with large intensity-attenuations are fitted to the equation [$y = \Sigma a_i\text{-}b_i^{-cx}$], where *i* represents nine individual energies (258, 569, 743, 786, 880, 883, 925, 927, and 946 keV). Note that the data of the two most intense gamma rays of ^{238}U, 766 and 1001 keV, are not used because those peaks have no intensity-attenuations and therefore would not contribute to calculating the intensity-attenuation coefficient *c*.

After the intensity-attenuation coefficient *c* is found, the individual measured-to-fit ratios of each energy peak are then fitted to the equation $y = a\text{-}be^{-cx}$, where *c* is now a known constant. The results of some of the gamma rays relevant to the isotopic analysis are presented in Table I. Because the results of the data sets of 0.7% and 4.5% ^{235}U without the absorber are similar and those with the absorbers are similar, they are combined into without-absorber and with-absorber

Table II. Comparison of gamma-ray emission probabilities with some other studies.

Isotope	Energy	Ref [2]	err	Ref [1]	err	Present work	err
^{234}U	120.90	3.342E-04	5.0E-06			3.466E-04	4.2E-06
^{235}U	143.76	1.096E-01	8.0E-04			1.095E-01	2.3E-04
^{235}U	163.33	5.080E-02	4.0E-04			5.089E-02	7.4E-05
^{235}U	185.72	5.720E-01	5.0E-03			5.737E-01	9.6E-04
^{235}U	194.94	6.300E-03	1.0E-04			6.266E-03	2.1E-05
^{235}U	202.11	1.080E-02	2.0E-04			1.094E-02	3.5E-05
^{235}U	205.31	5.010E-02	5.0E-04			4.993E-02	1.1E-04
^{238}U	258.26	7.280E-04	4.0E-06	7.300E-04	3.4E-06	7.228E-04	4.0E-06
^{238}U	568.80	5.770E-05	6.6E-06	N/A†	N/A	5.739E-05	3.1E-07
^{238}U	569.40	1.320E-04	1.3E-05	2.030E-04	2.6E-06	1.311E-04	7.2E-07
^{238}U	742.81	8.330E-04	4.0E-05	9.460E-04	6.6E-06	8.942E-04	3.4E-06
^{238}U	766.36	2.940E-03	1.2E-04	3.220E-03	2.1E-05	3.069E-03	1.1E-05
^{238}U	786.27	5.040E-04	1.9E-05	5.540E-04	5.2E-06	5.398E-04	1.8E-06
^{238}U	880.45	2.040E-04	N/A	2.120E-04	1.9E-06	2.103E-04	1.2E-06
^{238}U	883.22	1.880E-04	1.0E-05	2.110E-04	1.9E-06	2.108E-04	1.2E-06
^{238}U	945.96	3.130E-04	1.6E-05	3.350E-04	2.9E-06	3.438E-04	1.4E-06
^{238}U	1001.03	8.370E-03	1.0E-04	8.390E-03	4.7E-05	8.371E-03	2.9E-05

groups for the ease of comparison and inspection. Additionally, shown in this table are the results of the 121-keV peak of ^{234}U, which is also used for determining ^{234}U in the uranium isotopic analysis. The results of this peak were calculated the same way as those of ^{235}U and ^{238}U using the certified values of ^{234}U in the two sources.

The new BRs from both groups agree well within the uncertainty limits and can be combined in the final results. However, the fractional intensity-attenuations (*b/a*) from the group with absorbers are completely different from those without absorbers. In general, the fractional intensity-attenuations of the group with the absorber are somewhat lower than those of the group without the absorber. The reason is that the absorber would absorb some of the coincidence gamma rays thus lessening the chance for summing. The intensity-attenuation reduced by the cadmium absorber is different for different energy peaks due to the number and energies of the coincidence peaks. If the peak is in coincidence with some low-energy gamma rays or x-rays, then the addition of a thin cadmium absorber would absorb most of the coincident rays and reduce the summing effect significantly. However, if it is in coincidence with mostly high-energy gamma rays, then the introduction of a thin cadmium absorber would not affect the intensity-attenuation by much. For most of the isotopic analysis measurements in this energy range (such as those of FRAM), some cadmium or tin absorber is used to absorb the x-rays (to reduce the summed peaks). Therefore, it is suggested that only the fractional intensity-attenuation values of the set with cadmium absorber should be used.

The fractional intensity-attenuations shown in Table I are dependent on the geometry of the sources and detector. With differences in source size and mass, those values may be changed somewhat. For example, a very thin uranium foil will allow most of the gamma rays and x-rays to enter the detector, and this would significantly affect the fractional intensity-attenuation due to summing of many gamma rays in coincidence with low-energy rays. However, a large block of

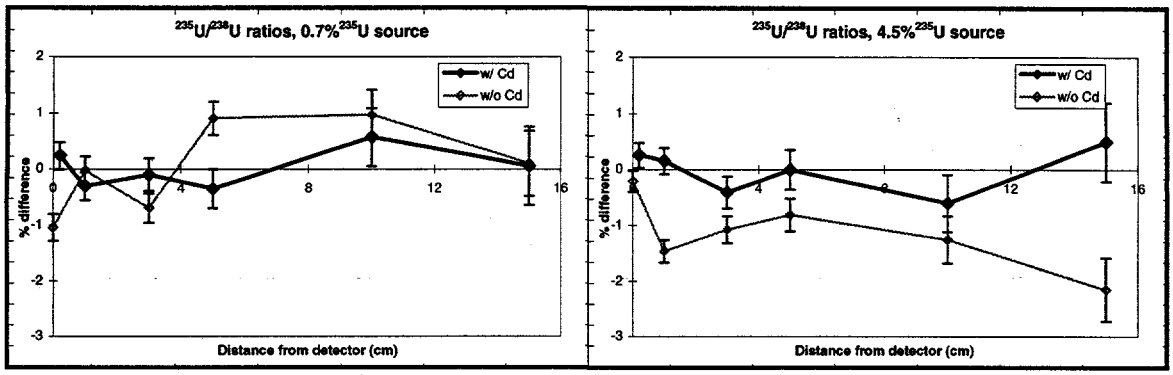

Fig. 5. Results of FRAM using the modified parameter file with the corrected BRs and the intensity-attenuation corrections. The vertical scales show the differences between the calculated ratios and the accepted ones.

uranium (on the order of hundreds or thousands of grams) will not affect the fractional intensity-attenuation shown here by much because in this experiment, the thicknesses of the sources were considered to be infinite for the gamma rays with energies of several hundred keV. It is fortunate that due to the nature of uranium isotopic determination which normally involves measuring a large quantity, the intensity-attenuation of the peaks will not significantly change from the present values.

With different detectors, the absolute intensity-attenuation values may also be changed, but the relative values (i.e., the ratios of the values) should remain the same. So, the intensity-attenuation correction values in Table I will also work with other detectors.

Table II shows the comparison of the BR results with some other studies. The results are the weighted averages of all four sets of data and may be slightly different from the averaged results of the two groups in Table I. The quoted errors of the present work include both the statistical and instrumental uncertainties of all four sets of data. For the ^{235}U, the results of the present work agree well with those of Ref. [2], and the quoted errors are much smaller. As for the ^{238}U, in general, the results of the present work fall between those of Refs. [1] and [2] and tend to bridge the large differences between those two references.

C. Results Using FRAM with Modified Parameter File

A new parameter was created using the new BRs of the present work to determine the isotopic ratios of uranium. The 766-, 786-, 880-, 883-, and 946-keV peaks were used to determine the intensity-attenuation correction factors for the other peaks of ^{238}U. The intensity-attenuation values used are those of the group with the cadmium and copper absorbers. Note that even though the absolute intensity-attenuation factors do change with different distance, the relative factors should remain the same regardless of the distances. Because of the way the FRAM code was written and because these peaks are used for intensity-attenuation correction, they can not be used for relative efficiency. Therefore, only the 144-, 163-, 186-, and 205-keV peaks of ^{235}U and the 258-, 569-, 743-, and 1001-keV peaks of ^{238}U and its daughters are used to determine the relative efficiencies. As with the old parameter file, only the 258-keV peak is used to determine the activity of ^{238}U.

The new parameter was used to determine the $^{235}U/^{238}U$ ratios for the data. It turned out that the averaged results were about 1.5% lower than expected. The reason for the discrepancies is not

due to the inaccuracies of the BRs or the intensity-attenuation, but comes from overestimating the intensity-attenuation correction factor from the current version of FRAM.

Because the 258-keV peak is the only one used in determining the activity of ^{238}U and is the main peak to determine the relative efficiencies to correct for the discrepancies of 1.5%, the BR of 258-keV peak is adjusted upward by 1.5% to a value of 7.336e-4. The results of all four sets of data (with and without the absorbers) using the new parameter with the adjusted 258-keV peak are shown in Fig. 5. As for the results of the group without the absorbers, because the intensity-attenuation correction factor was calculated using the results of the with-absorber-group, they are not expected to be correct. Those results are only shown for information and comparison with the results of the other group and not for the accuracy.

V. CONCLUSION

A new method of determining the BRs and the fractional intensity-attenuations of the peaks due to summing has been presented. The uranium results can be used to better determine the uranium isotopic ratios. Using the isotopic analysis code FRAM, even with the limitation of the current version, the results of the uranium isotopic ratios with the new BRs and fractional intensity-attenuation corrections have been shown to be much better than those with the old BRs and without any corrections for the intensity-attenuations of the peaks due to summing. In the future version of FRAM where some of the limitations are removed, the uranium isotopic analysis, when used with the findings in this work, would be even more accurate.

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* This peak is fixed to the 569.4-keV peak in the calculations.

† The BR of this peak is combined with the 569.4-keV peak.