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Generalization of the IDGS Technique

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ABSTRACT

Isotope Dilution Gamma-ray Spectrometry (IDGS) is a method using gamma-ray spectrometry technique to measure plutonium in solutions. To measure the plutonium concentration, the unknown solutions are spiked with plutonium of accurately known plutonium mass and isotopic composition. The isotopic compositions of the unknown solution and the spiked solution are determined by high-resolution gamma-ray measurements. In the traditional IDGS method, the concentration of plutonium in the unknown solution is determined by calculating the differences among the ^{239}Pu weight percent and isotopic ^{240}Pu/^{239}Pu ratios of the spike, the spiked solution, and the unknown solution.

To improve the measurement result, we are developing a generalization method for the IDGS technique. The generalized IDGS method utilizes all the plutonium isotopes (except ^{242}Pu) and americium fractions. This would allow the calculations of the concentration of plutonium in the unknown solution using other plutonium isotopic ratios in addition to the isotopic ^{240}Pu/^{239}Pu ratio.

I. INTRODUCTION

In the traditional IDGS method that has been employed to measure input dissolver solutions at Tokai Reprocessing plant in Japan, the concentration of plutonium in the unknown solution is then determined by calculating the differences among the isotopic ^{240}Pu/^{239}Pu ratios of the spike, the spiked solution, and the unknown solution.

$$\mathrm{C(Pu)} = \frac{M_s}{V_u} \bullet \frac{W_s^{9}}{W_u^{9}} \bullet \frac{R_m - R_s}{R_u - R_m}, \qquad (1)$$

where

M_s = mass of plutonium in the spike
V_u = volume of dissolver solution taken
W_s^9 = weight fraction of ^{239}Pu in the spike
W_u^9 = weight fraction of ^{239}Pu in the unknown solution
R_m = the ^{240}Pu/^{239}Pu ratio in the mixed or spiked solution
R_s = the ^{240}Pu/^{239}Pu ratio in the spike
R_u = the ^{240}Pu/^{239}Pu ratio in the unknown solution

In this equation, the values of M_s, V_u, W_s^9, and R_s are known. Only the values of R_u and W_u^9 in the unknown solution and R_m in the spiked solution are needed to be measured by gamma-ray spectrometry [1,2].

Equation 1 utilizes only the isotopic $^{240}Pu/^{239}Pu$ ratios of the spike, the spiked solution, and the unknown solution. To enhance the IDGS capability, we are developing a generalization of the IDGS technique that allows the calculations of the concentration of plutonium in the unknown solution using other plutonium isotopic ratios in addition to the isotopic $^{240}Pu/^{239}Pu$ ratio.

The generalized equation for the concentration of plutonium in the unknown solution then can be written as

$$\mathrm{C(Pu)}^{ij} = \frac{M_s}{V_u} \bullet \frac{W_s^{\,i}}{W_u^{\,i}} \bullet \frac{R_m^{ij} - R_s^{ij}}{R_u^{ij} - R_m^{ij}}, \qquad (2)$$

where

M_s	= mass of plutonium in the spike
V_u	= volume of dissolver solution taken
$W_s^{\,i}$	= weight fraction of isotope ***i*** in the spike
$W_u^{\,i}$	= weight fraction of isotope ***i*** in the unknown solution
$R_m^{\,ij}$	= the ratio of isotope ***j*** to isotope ***i*** in the spiked solution
$R_s^{\,ij}$	= the ratio of isotope ***j*** to isotope ***i*** in the spike
$R_u^{\,ij}$	= the ratio of isotope ***j*** to isotope ***i*** in the unknown solution
i, ***j***	= plutonium or americium isotopes (^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{241}Am)

This equation is (***i, j)*** symmetrical. That is,

$$\mathrm{C(Pu)}^{ij} = \frac{M_s}{V_u} \bullet \frac{W_s^{\,i}}{W_u^{\,i}} \bullet \frac{(R_m^{ij} - R_s^{ij})}{(R_u^{ij} - R_m^{ij})} = \frac{M_s}{V_u} \bullet \frac{W_s^{\,i}}{W_u^{\,i}} \bullet \frac{\left(1/R_m^{\,ji} - 1/R_s^{\,ji} \right)}{\left(1/R_u^{\,ji} - 1/R_m^{\,ji} \right)} = \frac{M_s}{V_u} \bullet \frac{W_s^{\,i}}{W_u^{\,i}} \bullet \frac{\left((R_s^{\,ji} - R_m^{\,ji}) / R_m^{\,ji} R_s^{\,ji} \right)}{\left((R_m^{\,ji} - R_u^{\,ji}) / R_u^{\,ji} R_m^{\,ji} \right)}$$

$$= \frac{M_s}{V_u} \bullet \frac{W_s^{\,i}}{W_u^{\,i}} \bullet \frac{R_u^{\,ji}}{R_s^{\,ji}} \bullet \frac{(R_m^{\,ji} - R_s^{\,ji})}{(R_u^{\,ji} - R_m^{\,ji})} = \frac{M_s}{V_u} \bullet \frac{W_s^{\,i}}{R_s^{\,ji}} \bullet \frac{R_u^{\,ji}}{W_u^{\,i}} \bullet \frac{(R_m^{\,ji} - R_s^{\,ji})}{(R_u^{\,ji} - R_m^{\,ji})} = \frac{M_s}{V_u} \bullet \frac{W_s^{\,j}}{W_u^{\,j}} \bullet \frac{(R_m^{\,ji} - R_s^{\,ji})}{(R_u^{\,ji} - R_m^{\,ji})} = \mathrm{C(Pu)}^{ji}$$

In some processes, the mass of the unknown solution is used instead of the volume. Then the plutonium concentration $C_M(Pu)$ will have the unit of grams (or mg) of plutonium per unit mass of the unknown solution instead of grams of plutonium per unit volume of the unknown solution.

II. AGED PRODUCT SOLUTION

To test Equation 2, we analyze the gamma-ray data taken at the Plutonium Fuel Center, Japan Nuclear Cycle Development Institute (JNC), Tokai, Japan, using FRAM v4 [3,4] with the parameter set UPu60_210Solution. (The parameter set indicates the analysis is for plutonium and uranium in solution with the analysis in the energy range from 60 to 210 keV.) The samples were analyzed by Isotope Dilution Mass Spectrometry (IDMS), so we can compare the IDGS results with the IDMS results. These data include ten spectra, five each of the unknown and spiked solutions, at five different measurement live times: 0.5, 1, 2, 3, and 4 hours. The isotopic compositions of the spike were already known and require no gamma measurements. Table 1 shows the plutonium isotopic compositions of the spike.

Table 1. Isotopic compositions of the spike. Pu/U ratio equals 0.048613.

Isotope	Percents
Pu238	0.00354
Pu239	97.76098
Pu240	2.20018
Pu241	0.02825
Pu242	0.00709
U234	0.00538
U235	0.71208
U236	0.00012
U238	99.28242

The isotopic compositions in Table 1 are already decay corrected to the time of the gamma-ray measurements. There is no ^{241}Am information. We, however, from the gamma-ray measurements of other spike samples, estimate it to be about 440 parts per million (ppm). This ^{241}Am composition will be used later in the IDGS calculations of the concentration of plutonium in the unknown sample.

We analyzed the ten spectra (5 of unknown solution and 5 of spiked solution). The results of those measurements are shown in Tables 2 and 3.

The mass spectrometry results are also shown in these two tables, so we can compare the gamma results with the mass spectrometry results.

Table 2. Isotopic compositions of the unknown solution. The ^{242}Pu is determined from the correlation. The ^{241}Am and ^{235}U mass are expressed as percents of the plutonium mass.

Spectra	Pu238		Pu239		Pu240		Pu241		Pu242		Am241		U235	
	Mass %	% err	Mass %	% err	Mass %	% err	Mass %	Mass %	Mass %	% err	Mass %	% err	Mass %	% err
IDMS	1.239		62.267		25.64		6.211		4.643				43.67	
4 h spec	1.205	1.70	61.870	0.61	25.924	1.26	5.972	0.93	5.028	2.32	3.121	1.00	40.46	3.79
3 h spec	1.145	2.02	61.886	0.71	26.136	1.46	5.914	1.08	4.919	2.69	3.096	1.15	40.40	4.40
2 h spec	1.221	2.38	61.869	0.85	25.895	1.73	5.962	1.30	5.053	3.24	3.095	1.39	39.98	5.35
1 h spec	1.266	3.45	62.668	1.26	25.027	2.72	6.090	1.94	4.949	4.83	3.196	2.07	44.85	7.15
0.5 h spec	1.339	4.75	60.685	1.87	25.997	3.70	6.416	2.73	5.563	6.86	3.333	2.90	43.40	11.1

Table 3. Isotopic compositions of the spiked solution. The ^{242}Pu is determined from the correlation. The ^{241}Am and ^{235}U mass are expressed as percents of the plutonium mass.

Spectra	Pu238		Pu239		Pu240		Pu241		Pu242		Am241		U235	
	Mass %	% err	Mass %	% err	Mass %	% err	Mass %	% err	Mass %	% err	Mass %	% err	Mass %	% err
IDMS	0.415		85.953		9.998		2.085		1.549				24.30	
4 h spec	0.401	3.09	86.787	0.28	10.110	2.28	2.043	1.25	0.659	2.91	1.073	1.42	24.03	5.04
3 h spec	0.389	3.61	86.720	0.32	10.225	2.55	2.010	1.44	0.655	3.33	1.095	1.62	21.32	6.42
2 h spec	0.406	4.35	86.087	0.40	10.784	3.03	2.021	1.77	0.702	4.04	1.048	2.01	23.65	7.18
1 h spec	0.409	5.97	86.332	0.56	10.517	4.28	2.051	2.48	0.692	5.64	1.081	2.80	23.64	10.3
0.5 h spec	0.456	7.99	87.455	0.76	9.440	6.62	1.990	3.57	0.658	8.12	1.094	4.02	20.03	15.9

The ^{242}Pu values in Table 3 are determined with isotopic correlation. The isotopic correlation in FRAM works reasonably well for plutonium material from various types of reactors but not for mixed plutonium as in this case where very low burn-up plutonium is mixed with high burn-up plutonium. This inability to accurately determine ^{242}Pu in the mixed plutonium solution would not hamper the determination of the plutonium concentration because, as we see from Equations 1 and

2, all the terms involving mixed or spiked solutions are the isotopic ratios and they are independent of the ^{242}Pu fraction in the solution.

Note that the percent errors shown in Tables 2 and 3 are those of the isotopic fraction of the individual isotopes after all the isotopic correlations are taken into account. Equation 2 includes the ratios of two isotopes. In the error propagation of the uncertainty of the plutonium concentration, the errors of all these ratios would need to be accounted for. The errors of the isotopic ratios would be well overestimated if they were calculated from the final, correlated errors of individual isotope fractions in Tables 2 and 3. In fact, they should be calculated from the uncorrelated errors of the individual isotopes. Those uncorrelated errors are available in the "results of activity calculations" section in the medium or long output of the isotopic analysis code FRAM. Table 4 shows an example of the isotopic ratios and uncorrelated errors of the isotopes.

Table 4. Isotopic ratios of the 4-h spectrum of the unknown solution. The isotope % errors in the fourth column are the uncorrelated errors of the individual isotopes, not of the ratios of two isotopes.

Isotopic ratios	Activity ratios	Mass ratios	Isotope % errors
Pu239/Pu239	1.00E+00	1.00E+00	1.31%
Pu241/Pu239	1.61E+02	9.65E-02	0.30%
Am241/Pu239	2.78E+00	5.04E-02	0.47%
Pu238/Pu239	5.38E+00	1.95E-02	1.47%
Pu240/Pu239	1.53E+00	4.19E-01	1.29%
Pu242/Pu239	0.00E+00	0.00E+00	>99.99%
U235/Pu239	2.28E-05	6.54E-01	3.74%

The percent errors in the fourth column in Table 4 show the uncorrelated errors of the individual isotopes (the numerators of the isotopic ratios in column 1) and not the errors of the isotopic ratios. ^{242}Pu does not have any measurable gamma rays, so its activity or mass ratios are shown as zeros in Table 4. Its final fractional results will be determined from correlation or by operator input.

Table 5 shows the ratios of the plutonium concentrations C_M(Pu) determined from the 4-h spectra (unknown and spiked solutions) using Equation 2 to that determined by IDMS. The errors of M_s, M_u (mass of unknown solution instead of volume V_u), W_s^i, and R_s^{ij} are assumed to be negligible (compared with those from gamma-ray measurements) and are ignored in the error propagations.

Table 5. Ratios of the plutonium concentrations C_M(Pu) determined from the 4-h spectra using Equation 2 to that of the concentration determined from IDMS, which was 0.9545 mg/g. The sum results in column 7 are the weighted average of those in columns 2-6. The results at the bottom of column 7 are the weighted average off all the results.

Isotope *j* / Isotope *i*	Pu238		Pu239		Pu240		Pu241		Am241		Average	
	Conc	% err	Conc	% err	Conc	% err	Conc	% err	Conc	% err	Conc	% err
Pu238			0.977	4.5	0.945	27.2	0.118	116.8	0.642	100.9	0.974	4.45
Pu239	0.977	4.3			0.985	4.2	1.010	2.4	0.992	2.6	0.996	1.51
Pu240	0.945	25.2	0.985	3.9			1.140	19.8	1.033	21.6	0.991	3.70
Pu241	0.118	115.0	1.010	2.1	1.140	20.9			3.458	219.2	1.011	2.09
Am241	0.642	99.0	0.992	2.3	1.033	22.7	3.458	218.5			0.993	2.31
All Isotopes											**0.998**	**1.43**

We can see that the results are symmetrical through the diagonal as expected. That is, $C_M(Pu)^{ij}$ are the same as $C_M(Pu)^{ji}$. The errors are also almost symmetrical. The small differences are due to

rounded off and correlation calculations of the errors. Column 7 shows the weighted average of the results in columns 2-6. The results at the bottom of column 7 are the weighted average off all the results from either side of the diagonal.

The calculations with ^{239}Pu appear to give the best results of all. The reason is not due to the large fractions of ^{239}Pu in the spike or the solutions or the small uncertainties of the ^{239}Pu isotope (see Table 4); the reason is due to the large differences of the isotopic ratios (where ^{239}Pu is in the denominator) between the spike, spiked solution, and unknown solution. In fact, the average of the six results involving non-^{239}Pu isotopes (^{238}Pu, ^{240}Pu, ^{241}Pu, and ^{241}Am) comes out to be 1.05 with 13% uncertainty. This uncertainty is about a factor of ten larger than the average results of those involving ^{239}Pu. This means that even though we can use the ratios of all the isotopes to determine the concentration of plutonium in the solution, we do not need to. We only need the ratios of all the isotopes to that of ^{239}Pu in the calculations, and the result will be almost the same as if all the isotopes have been used.

Table 6 shows the ratios of the plutonium concentrations C_M(Pu) determined from all five sets of spectra using Equation 2 where ***i*** is ^{239}Pu and ***j*** is the other four isotopes to that of the IDMS.

Table 6. Ratios of the plutonium concentrations CM(Pu) determined from the five sets of spectra using Equation 2 with isotope *i* fixed to ^{239}Pu to that of the IDMS, which was 0.9545 mg/g. The sum results in column 6 are the weighted average of those in columns 2-5.

Isotope *j* / Spectra	Pu238		Pu240		Pu241		Am241		Average	
	Conc	% err	Conc	% err	Conc	% err	Conc	% err	Conc	% err
4 h spec	0.9770	4.26	0.9847	4.23	1.0099	2.39	0.9923	2.57	0.9964	1.51
3 h spec	1.0074	5.01	0.9930	4.79	1.0020	2.76	1.0315	2.98	1.0116	1.75
2 h spec	0.9877	6.00	1.1105	5.78	1.0086	3.38	0.9817	3.63	1.0105	2.13
1 h spec	0.9505	8.40	1.1196	8.53	1.0000	4.88	0.9813	5.22	1.0024	3.06
0.5 h spec	0.9929	11.50	0.8616	12.23	0.8749	6.85	0.9185	7.38	0.9046	4.31

We see that most of the results agree with the accepted concentration from IDMS determination within the uncertainties. The results and uncertainties behave well with respect to the acquisition time; that is, the uncertainty is proportional to the square root of the acquisition time. Also, for the individual isotopic ratio, ^{241}Pu/^{239}Pu (and its inverse) would give the best result. The best results of all would be the average, which is almost a factor of three better than that using the ^{240}Pu/^{239}Pu ratio as in the traditional IDGS method. This also means that if we use the average results, then we can in half an hour obtain the results equivalent to 4 hours of acquisition with the traditional IDGS calculations (with ^{239}Pu for isotope ***i*** and ^{240}Pu for isotope ***j***).

Note that these isotopes are not restricted to only plutonium isotopes but any isotopes such as ^{241}Am (shown in Tables 5 and 6) or uranium isotopes. This means that if we can determine the uranium isotope fractions relative to plutonium, then we can also use those ratios in the calculations of the plutonium concentration. For these spectra, the activities of ^{238}U are very weak and cannot be accurately determined. We determined the ^{235}U ratios to plutonium. We, however, did not include ^{235}U in these calculations because they would not contribute much to the final average results due to the relatively large uncertainties of ^{235}U in these spectra (see Table 4). It is mentioned here, so we know that ^{235}U can be used in the calculations of plutonium concentration and should be used if its relative uncertainty is reasonably small or comparable to the uncertainties of other isotopes.

III. FRESHLY SEPARATED SOLUTION

The aged plutonium has a huge 60-keV peak from ^{241}Am decay, and that peak and its Compton would overwhelm all the peaks below it, making them useless for isotopic measurements. For freshly separated plutonium, the 60-keV peak is small, and the peaks below it can be used for isotopic determination. These peaks below 60-keV are intense, and the analysis employing this region may give better isotopic results than the region above 60-keV, which then would lead to better IDGS determination.

For that reason, we tested the IDGS analysis with the freshly separated plutonium at the Tokai Plutonium Fuel Center, JNC, in order to improve the IDGS technique. For freshly separated product solution or spent-fuel dissolver solutions, because plutonium, uranium, and americium concentrations are altered differently during the separation process, we will not be able to use uranium or americium, but only plutonium, to determine the plutonium concentration. That means we can only use the isotopic ratios of ^{238}Pu/^{239}Pu, ^{240}Pu/^{239}Pu, and ^{241}Pu/^{239}Pu in the calculations of the plutonium concentration.

Two sets of freshly separated plutonium data, one for the unknown solution and one for the spiked solution, were obtained. Twenty-four spectra were acquired for each set accumulatively in a twenty-four hour period. The samples were analyzed by IDMS, so we can compare the IDGS results with the IDMS results. Tables 7 and 8 show the plutonium isotopic compositions of the unknown and spiked solutions analyzed by FRAM with the parameter set Upu38_186SolFresh. Twenty-four spectra were obtained for each set, but we only show those at 1, 2, 4, 8, 16, and 24 hours acquisition time.

Comparing the results in these two tables with those in Tables 2 and 3, we see that the reported errors of ^{238}Pu, ^{239}Pu, and ^{240}Pu of the fresh plutonium samples are just only slightly worse than those of the aged samples for the same acquisition times. This is probably due to the amount of material to measure, which was less than that of the aged plutonium measurements. Note that the amounts of plutonium involved in the separations were originally about the same as the amounts used in the aged plutonium measurements and that the plutonium recovery rate from the ion exchange is about 70%.

Table 7. Isotopic compositions of the unknown solution. The ^{242}Pu is determined from the correlation, which was adjusted to better match that of the IDMS.

Spectra	Pu238		Pu239		Pu240		Pu241		Pu242	
	Mass %	% err	Mass %	% err	Mass %	% err	Mass %	Mass %	Mass %	% err
IDMS	0.576		70.731		23.273		3.043		2.378	
1 h spec	0.583	4.67	71.326	1.58	22.593	4.36	3.151	6.70	2.347	7.69
2 h spec	0.578	3.26	71.696	1.08	22.120	3.05	3.298	4.50	2.308	5.34
4 h spec	0.593	2.29	70.748	0.79	22.992	2.13	3.227	3.17	2.441	3.79
8 h spec	0.589	1.59	70.792	0.55	23.081	1.48	3.121	2.22	2.418	2.63
16 h spec	0.582	1.12	70.940	0.38	23.013	1.04	3.083	1.55	2.383	1.85
24 h spec	0.577	0.91	70.974	0.31	22.978	0.84	3.100	1.26	2.372	1.49

Table 8. Isotopic compositions of the spiked solution. The ^{242}Pu is fixed to that of the IDMS measurement.

Spectra	Pu238 Mass %	Pu238 % err	Pu239 Mass %	Pu239 % err	Pu240 Mass %	Pu240 % err	Pu241 Mass %	Pu241 % err	Pu242 Mass %	Pu242 % err
IDMS	0.192		88.847		9.150		1.022		0.789	
1 h spec	0.194	5.43	89.419	0.58	8.562	5.53	1.037	8.71	0.789	0.01
2 h spec	0.200	3.76	89.015	0.42	8.997	3.81	1.000	6.17	0.789	0.01
4 h spec	0.199	2.65	88.966	0.29	9.003	2.68	1.044	4.30	0.789	0.01
8 h spec	0.191	1.87	89.022	0.21	8.945	1.88	1.052	3.02	0.789	0.01
16 h spec	0.192	1.32	88.984	0.15	8.998	1.33	1.038	2.10	0.789	0.01
24 h spec	0.192	1.07	88.882	0.12	9.103	1.08	1.034	1.72	0.789	0.01

The ^{241}Pu errors are about a factor of 3.5 larger than those of aged plutonium for the same acquisition times. The large difference is mainly due to the use of the ^{237}U peaks for the measurements of the ^{241}Pu in aged plutonium and not in fresh plutonium.

Table 9 shows the ratios of the plutonium concentrations C_M(Pu) determined using Equation 2 where ***i*** is ^{239}Pu and ***j*** is ^{238}Pu, ^{240}Pu, and ^{241}Pu to that of the IDMS.

Table 9. Ratios of the plutonium concentrations CM(Pu) determined from the five sets of spectra using Equation 2 with isotope *i* fixed to ^{239}Pu to that of the IDMS, which was 0.6040 mg/g. The sum results in column 5 are the weighted average of those in columns 2-4.

Isotope *j* / Spectra	Pu238 Conc	Pu238 % err	Pu240 Conc	Pu240 % err	Pu241 Conc	Pu241 % err	Average Conc	Average % err
1 h spec	0.9869	9.88	0.9230	12.01	0.9679	14.14	0.9627	6.72
2 h spec	1.0514	6.97	1.0494	8.49	0.8732	9.62	1.0083	4.70
4 h spec	1.0035	4.86	0.9874	5.89	0.9497	6.84	0.9861	3.29
8 h spec	0.9602	3.37	0.9702	4.11	1.0040	4.83	0.9732	2.29
16 h spec	0.9813	2.38	0.9860	2.89	1.0028	3.37	0.9877	1.61
24 h spec	0.9987	1.94	1.0105	2.36	0.9916	2.74	1.0007	1.31

The average results are almost a factor of two better than that of the one using only the ^{240}Pu/^{239}Pu ratio of the traditional IDGS method. Comparing the results in Table 9 with the results in Table 6, we see that the results from the calculations of individual isotopic ratios, ^{238}Pu/^{239}Pu, ^{240}Pu/^{239}Pu, and ^{241}Pu/^{239}Pu of the fresh plutonium samples are somewhat worse than those of the aged samples and that the average results are about a factor of two worse.

IV. DISCUSSION

A. Systematic errors from the correlation calculations of ^{242}Pu

Since ^{242}Pu cannot be determined directly from gamma-ray measurements, we do not use it in the calculations of the plutonium concentration. However, even though it is not used directly in the calculations, it can affect the plutonium concentration result and its error indirectly. Equation 2 has the weight fraction of isotope ***i*** in the unknown solution term W_u^i. A 10% error for ^{242}Pu in the

unknown solution, which is not unreasonable since ^{242}Pu is determined from correlation, can translate into about 0.5% errors in all other isotopes.

This error caused by ^{242}Pu is not random but systematic. It depends on the correlation and on the material type or the type of the reactors that the plutonium comes from. This error can be reduced if the ^{242}Pu correlation is tuned up for the materials from different reactors. That is, we will have a number of similar FRAM parameter sets; each one differs from the others by just a constant in the correlation formula. The ^{242}Pu correlation for each of these parameter sets is tuned up specifically for the plutonium from a type of reactor (example BWR, PWR, CANDU, etc.) or reactor burning different fresh fuel (uranium or MOX). Then the uncertainty of ^{242}Pu may be reduced to perhaps below 4%, which translates to less than 0.2% systematic uncertainty of W_u^i caused by ^{242}Pu.

B. IDGS improvements

The results of the aged samples in Section 2 and fresh samples in Section 3 show that it is better to analyze the aged samples than going through the ion exchange process to get a result that is a factor of two less accurate. The accuracies of the results in these analyses are limited by the statistics. If better accuracy were designed, then the statistics would need to be improved by using more plutonium in the measurements, acquiring the data for a longer time, or using a higher efficiency detector.

Also, the average results from this generalized IDGS technique are almost a factor of three and two better than those of the traditional IDGS for the aged and fresh plutonium, respectively.

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