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ADVANCED ANALYSIS TECHNIQUES FOR URANIUM ASSAY

W.H. Geist, N. Ensslin, and L.A. Carrillo
Los Alamos National Laboratory, MS E540, Los Alamos, NM 87545

C.A. Beard
University of Texas at Austin, Austin, TX 78712

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ABSTRACT

Uranium has a negligible passive neutron emission rate making its assay practicable only with an active interrogation method. The active interrogation uses external neutron sources to induce fission events in the uranium in order to determine the mass. This technique requires careful calibration with standards that are representative of the items to be assayed. The samples to be measured are not always well represented by the available standards which often leads to large biases. A technique of active multiplicity counting is being developed to reduce some of these assay difficulties.

Active multiplicity counting uses the measured doubles and triples count rates to determine the neutron multiplication (M) and the product of the source-sample coupling (C) and the ^{235}U mass (m). Since the ^{235}U mass always appears in the multiplicity equations as the product of Cm, the coupling needs to be determined before the mass can be known. A relationship has been developed that relates the coupling to the neutron multiplication. The relationship is based on both an analytical derivation and also on empirical observations. To determine a scaling constant present in this relationship, known standards must be used. Evaluation of experimental data revealed an improvement over the traditional calibration curve analysis method of fitting the doubles count rate to the ^{235}U mass. Active multiplicity assay appears to relax the requirement that the calibration standards and unknown items have the same chemical form and geometry.

INTRODUCTION

Neutron coincidence counting is a standard technique used in the measurement and safeguarding of nuclear materials. These measurements involve counting the time-correlated neutrons to identify the fission neutrons originating from the special nuclear material. The rate of detection of fission neutrons is proportional to the ^{235}U mass. For the assay of uranium the technique of active coincidence counting must be used because the spontaneous fission rate is very small, making a passive measurement impracticable. The technique uses external neutron sources, often AmLi, to induce fission events in the sample. To determine the assay mass, the coincidence neutrons are counted in a similar way to passive assay of plutonium. One limitation of active coincidence counting is that the external neutron source used to induce fission events in the sample also results in accidental coincident events that limit the assay precision. Active neutron counters are designed to minimize the impact of the interrogation neutron sources on the assay [1].

The calibration curve approach is the traditional analysis method used to determine an assay mass for active coincidence counting. Using standards, a calibration curve is made that relates the ^{235}U mass to the doubles count rate. Certain requirements must be met for the calibration curve approach to result in a valid assay value, otherwise large biases can result. Some of these measurement requirements can be relaxed with the development of an analysis technique called active multiplicity analysis. This technique uses both the doubles and triples rates to solve for the assay mass and the sample multiplication.

The paper will discuss the ongoing work on the development of active multiplicity analysis techniques. For completeness, the traditional calibration curve approach will first be reviewed. The advantages and disadvantages of this technique will be discussed. We will then discuss the active multiplicity algorithm. The measurement of an experimental data set will be described. Both the calibration curve and active multiplicity analysis approaches will be applied to this data set and the results will be compared.

CALIBRATION CURVE APPROACH

The calibration curve approach works extremely well for many situations. A set of known standards is needed to calibrate the instrument. A nonlinear relationship between the doubles count rate and the ^{235}U mass is used as the calibration curve. The self-multiplication and coupling affect the shape of the calibration curve. The coupling is defined as the average number of fissions induced by the interrogation source per gram of ^{235}U in the sample. As the sample mass increases, the multiplication will tend to increase. This will cause the doubles rate to increase proportionally faster than the mass increase. The coupling will have an opposite effect on the doubles rate. As the mass increases the coupling decreases because the uranium located in the inner part of the object is shielded from the interrogation neutrons by the mass surrounding it. Often these two effects will tend to cancel one another, making the calibration curve approximately linear.

To obtain accurate results with the calibration curve approach, the calibration standards must be representative of the assay items. Some of the important sample characteristics are the enrichment, density, geometry, and material composition. The measurement bias will increase as the characteristics of the calibration standards and assay items diverge.

ACTIVE MULTIPLICITY APPROACH

Active multiplicity analysis has been under development at Los Alamos for many years [2-5]. This technique is similar to passive multiplicity analysis [6] that uses the singles, doubles, and triples count rates to solve for the sample mass, multiplication, and alpha value. In active multiplicity analysis, the doubles and triples count rates are solved for the sample multiplication and the product of the mass times coupling. The singles rate is not used because of the large background from the AmLi interrogation sources. The determination of the sample mass is not straightforward since the mass and the coupling always appear as a product in the active multiplicity equations.

Previous work on active multiplicity methods suggested that there was an empirical relationship between the coupling and the multiplication [3]. The coupling was found to

be inversely related to the multiplication and was found to be nearly independent of material composition. Recently a mathematical relationship was developed that relates the coupling to the multiplication and solid angle. This relationship can be written as

$$C = k\varphi(\Omega)\varphi(M),$$

where $\varphi(\Omega)$ represents the dependence of the coupling on the source-sample geometry, $\varphi(M)$ represents the dependence of the coupling on the flux depression within the sample, and k is a scaling factor. From this equation, the coupling is dependent on the solid angle, the multiplication, and a constant.

To understand the relationship between coupling and multiplication first consider the relationship that describes the probability of a fission neutron leaking from the sample to the probability it will induce further fission,

$$= \frac{\nu M}{M-1} - 1,$$

where ν is the average number of neutrons emitted per fission. This equation assumes that leakage is the main loss mechanism. If fission is the main absorption mechanism in the sample, then the flux depression of the AmLi interrogation neutrons will be the ratio of the probability that the neutrons will pass through the sample to the probability that they will be absorbed in fission. The functional form of these two cases is similar and the relationship for the coupling can be written as,

$$C = k\varphi(\Omega)\left(\frac{\nu M}{M-1} - 1\right).$$

Once the coupling is determined, the mass can be separated from the coupling in the multiplicity equations. The scaling constant, k, is determined from calibration standards.

EXPERIMENTAL DATA SET

To evaluate the active multiplicity analysis, experimental data was taken in a controlled setting. An active well coincidence counter (AWCC) [7] was used to measure uranium metal in several different configurations. The AWCC has two AmLi interrogation sources to induce fission events in the uranium samples. The uranium metal samples were in the form of metal pucks and several were used for each configuration. Three measurements were taken with a different number of pucks (different mass) in compact geometry for use as calibration points. The calibration configurations were chosen because the uranium mass was located closest to the same position for all three configurations. The rest of the measurements were performed with the pucks arranged in different configurations using aluminum metal spacers. The configurations are shown in Figure 1 along with the spacing dimensions. The count times ranged from 5 to 10 minutes which generally resulted in a precision in the triples count rate of better than 3%.

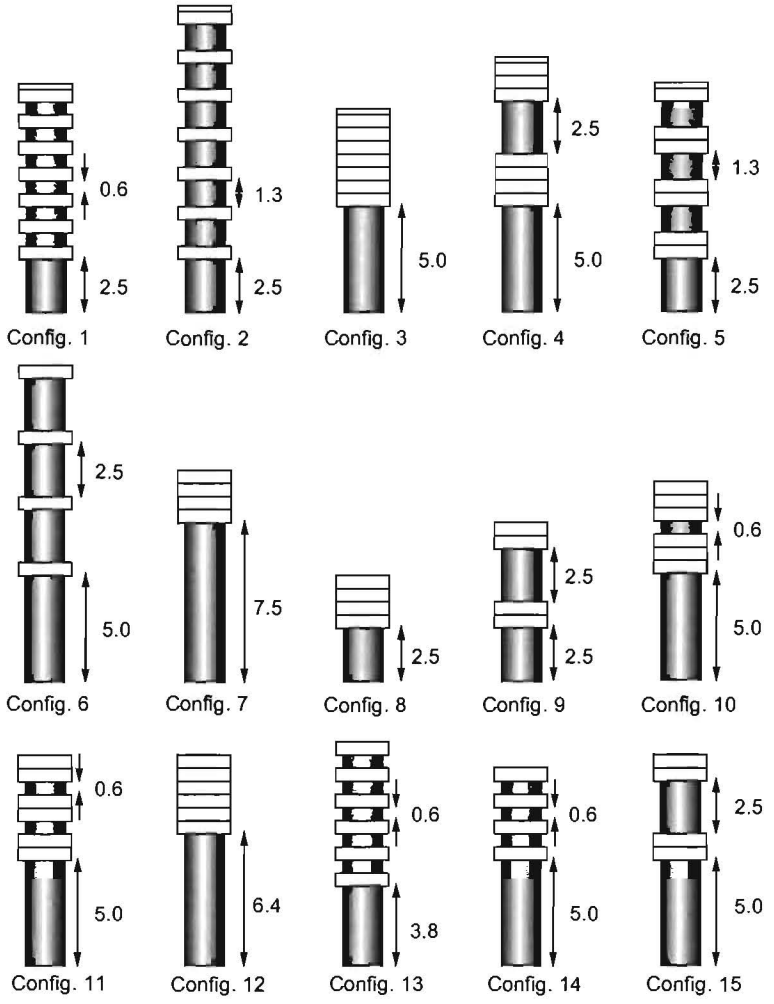

Figure 1. The 15 different configurations in which data was obtained. The uranium pucks are represented by the rectangles and the cylindrical looking shapes show the aluminum spacers. The dimensions of the height of the spacers are given in units of cm. Each uranium puck has a mass of approximately 528 grams except for one puck, which has about half the mass. The small mass puck is represented by the thinner rectangle. The configurations labeled 3, 7, and 12 were used as the calibration configurations.

ANALYSIS OF EXPERIMENTAL DATA SET

The data were analyzed with two techniques, the calibration curve approach and the active multiplicity approach. The calibration curve approach uses the doubles rate to determine the ^{235}U mass and the active multiplicity approach uses both the doubles and triples rates to determine the ^{235}U mass.

The relationship between the doubles count rate and the ^{235}U mass was determined from the three calibration configurations and is shown in Figure 2. The polynomial expression that describes this relationship is given by,

$$D = -46.196 + 0.138m + 6.6101\times10^{-7}m^2,$$

where D is the doubles rate and m is the ^{235}U mass.

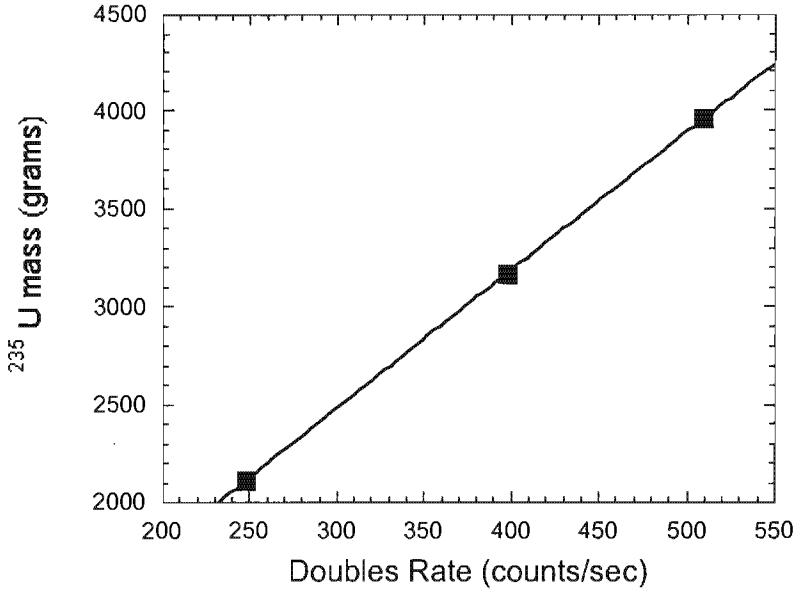

Figure 2. Calibration of the doubles count rate versus the uranium mass.

The remaining 12 configurations were analyzed using the relationship derived from the calibration curve. The results are shown in Table 1. The assay mass was biased low by an average of 6.5% relative to the known mass, and the data have a 1 sigma relative standard deviation scatter of 4.4%.

TABLE 1. Results of the calibration curve approach. Tabulated is the known mass, doubles rate, assay mass, and the error relative to the known mass.

Configuration	known mass	D	assay mass	% Error
1	3956	445.3	3508	-11.3
2	3956	460.9	3618	-8.5
4	3956	472.5	3699	-6.5
5	3956	453.3	3564	-9.9
6	2111	236.0	2028	-3.9
8	2111	257.8	2184	3.4
9	2111	238.3	2045	-3.1
10	3168	379.4	3045	-3.9
11	3168	370.3	2980	-5.9
13	3168	353.8	2864	-9.6
14	2111	223.9	1942	-8.0
15	2111	231.1	1994	-5.6

The analysis for active multiplicity counting is more complex than the calibration curve approach. The three calibration data points are used to solve for the scaling constant in the coupling relationship shown previously,

$$C = k\varphi(\Omega)\varphi(M)\,.$$

The solid angle factor, $\varphi(\Omega)$, is assumed to be constant and the actual scaling factor derived from the calibration standards is $k\varphi(\Omega)$. The scaling factor is derived from each standard by first determining the actual coupling defined as

$$C = F/g,$$

where F is the fission rate and g is the grams ^{235}U. The fission rate is determined from the formula

$$F = \frac{2D}{f\varepsilon^2 \nu_{s2} K},$$

where D is the doubles rate, f is the gate fraction, ε is the neutron detection efficiency, ν_{s2} is the second reduced factorial moment of the AmLi neutron induced fissions, and K is a correction factor that is determined from the multiplication using

$$K = M^2\left[1 + \frac{(M-1)\nu_{s2}\nu_{i2}}{\nu_{s2}(\nu_{i1}-1)}\right],$$

where ν_{s1} is the first reduced factorial moment of the AmLi neutron induced fissions and ν_{i1} and ν_{i2} are the first and second reduced factorial moments of the fission neutron induced fissions. The multiplication is determined from the active multiplicity equations using the measured the doubles and triples count rates [2]. Once the coupling is determined, the scaling constant is calculated by dividing the coupling by the $\varphi(M)$ term.

The scaling factor for the active multiplicity analysis was determined from the three calibration configurations and the results are shown in Table 2. The average scaling factor determined from calibration configurations is 0.0430. Note that although the fission rate and the coupling vary by several percent, the scaling constant only varies by 1% among the three calibration configurations.

TABLE 2. Data for the three active multiplicity calibration configurations. Tabulated is the known mass, fission rate, coupling, $\varphi(M)$, and the scaling constant.

Configuration	Mass	F	C	$\varphi(M)$	Scaling Constant
3	3956	1031.9	0.26	6.13	0.0425
7	2111	705.1	0.33	7.68	0.0435
12	3168	879.2	0.28	6.47	0.0429

Using this scaling factor determined from the calibration configurations, the mass of the other configurations can be solved for using the equations defined above. The multiplication and the product of the coupling times mass is determined from the active multiplicity equations. Then the coupling is determined using the multiplication and the scaling factor. The results are shown in Table 3. The mass was biased low by an average of 0.3% and the data have a 1 sigma relative standard deviation scatter of 2.9%.

The active multiplicity results are in better agreement with the known mass than the calibration curve results. A comparison between the two analysis types is shown in

Figure 3. The average bias was reduced from 6.5% to 0.3% when the data were analyzed with the active multiplicity analysis.

TABLE 3. The active multiplicity analysis for the assay configurations. Tabulated is the known mass, multiplication, $\varphi(M)$ factor, coupling, assay mass, and the error relative to the known mass.

Configuration	Known mass	M	$\varphi(M)$	C	Assay mass	% error
1	3956	1.41	8.10	0.34	3900.0	-1.42
2	3956	1.34	9.46	0.41	4137.5	4.59
4	3956	1.47	7.23	0.31	3985.1	0.74
5	3956	1.43	7.77	0.33	3924.9	-0.79
6	2111	1.23	12.89	0.55	2060.6	-2.39
8	2111	1.42	7.85	0.34	2238.8	6.06
9	2111	1.34	9.39	0.40	2138.2	1.29
10	3168	1.52	6.74	0.29	3095.8	-2.28
11	3168	1.47	7.27	0.31	3129.2	-1.22
13	3168	1.38	8.57	0.37	3138.3	-0.94
14	2111	1.33	9.64	0.41	2011.9	-4.69
15	2111	1.31	10.09	0.43	2078.5	-1.54

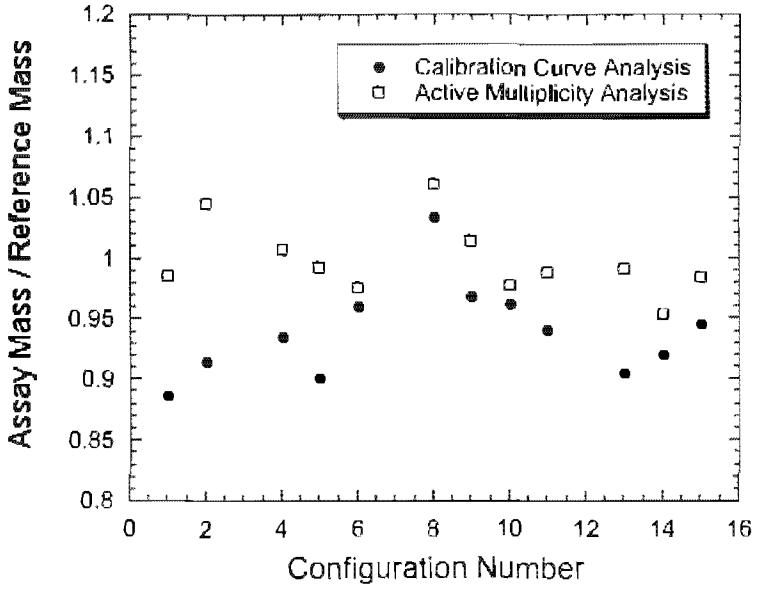

Figure 3. Comparison between the calibration curve analysis results (solid circles) and the active multiplicity analysis results (open squares) for the different configurations. An unbiased result will have an Assay Mass / Reference Mass value of unity.

RESULTS AND CONCLUSIONS

Active measurements were taken on different configurations of uranium disks to produce a data set that was used to compare two analysis techniques, calibration curve analysis and active multiplicity analysis. The calibration curve approach resulted in an average bias of 6.5% low relative to the known mass. This bias is likely the result of the calibration configurations differing from the other configurations. The active multiplicity approach resulted in better assay values with an average bias of only 0.3%. This improved assay is the result of using both the doubles and triples count rate to solve for

the multiplication. The multiplication is dependent on the uranium material configuration and this results in less dependence on the geometry for the active multiplicity analysis.

Continued development of active multiplicity techniques needs to be done. The current work used the same material form for both the determination of the scaling factor and the assay measurements. Experimental investigations to determine the sensitivity of the scaling factor to other materials forms need to be performed. Also, in the current work the solid angle factor, $\varphi(\Omega)$, was included in the scaling factor as a constant. The current results suggest that this factor is nearly constant, but additional investigation of the magnitude of this term needs to be undertaken. If the geometry of the assay items is constant, for example, the same size assay item is placed in the same position, then this factor will be nearly constant.

The successful development and implementation of active multiplicity techniques can have a beneficial impact on facility operations. Cost associated with standard development will be reduced and the calibration and operation of active counters will be simplified. Assay biases associated with poorly defined items and samples that lack representative calibration standards will be reduced.

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