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## NON-DESTRUCTIVE ANALYSIS OF IMPURE HEU-CARBON SAMPLES USING AN AWCC<sup>a</sup>

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### ABSTRACT

Highly enriched uranium-containing graphite-based material from the Los Alamos National Laboratory (LANL) is currently stored at the Idaho National Engineering and Environmental Laboratory (INEEL). Measurements to verify the uranium content of these samples are required prior to their disposition to the Y-12 facility in Tennessee. The stored materials vary significantly in their matrix purity and in their <sup>235</sup>U content and enrichment. A set of 26 samples selected from the LANL material inventory were analyzed non-destructively using an Active Well Coincidence Counter (AWCC) calibrated versus pure UO<sub>3</sub> standards. A correction, calculated from published data and the approximate carbon-to-uranium atom ratios of each sample, was applied for the response enhancement from the carbon matrix. In some cases this correction was as high as 30%. Eight of the 26 sample that had been analyzed in the AWCC were destructively analyzed to provide a benchmark for the non-destructive analyses. The average recovery (NDA/Destructive results) was 0.997±0.115. One sample had a destructive result that lay outside a 3-sigma interval about the NDA result.

### BACKGROUND

Highly enriched uranium (HEU)-containing graphite scrap materials from ROVER fuel operations at LANL in the 1960's are currently stored at the INEEL. These materials were received from LANL in 1986 through 1988. The INEEL is planning shipment of these materials to the Y-12 facility for final disposition. Prior to shipment the <sup>235</sup>U content must be verified. We have analyzed selected items from this inventory both nondestructively using an AWCC and destructively.

The LANL material is packaged in one-liter, two-liter or one-half gallon Nalgene<sup>®</sup> bottles and consists of three major categories:

1. Pyrolytic Carbon-U (PyC-U)	90 items,	1277 - 2204 g U,	92.6-93.2 % <sup>235</sup> U
2. Rotary Calciner Product (RCP)	131 items,	11 - 961 g U,	27.7-93.5 % <sup>235</sup> U
3. Rag Ash (RA)	54 items,	29 - 1516 g U,	34.9-97.6 % <sup>235</sup> U

Some items from each category were visually inspected. The PyC-U scrap is composed of pyrolytic carbon-encapsulated HEU in the form of very small almost powder-like beads. The Rotary Calciner Product (RCP) is a reasonably homogenous powdery material. The items in the Rag Ash (RA) category are more granular. Twenty-six items were selected from the LANL scrap inventory for testing and AWCC method development. Included were 6 RCP, 12PyC-U, and 8 RA samples.

The INEEL has a set of carefully calibrated UO<sub>3</sub> standards that were fabricated at Oak Ridge and analyzed both by Oak Ridge and by the New Brunswick Laboratory. However, we have no

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standards of HEU diluted with carbon. Dilution of the fissile material in samples with carbon has a pronounced effect on the response of an AWCC.<sup>1</sup> We lacked the resources to develop standards for HEU in carbon, so we relied on a correction technique similar to, but different than that outlined in the literature,<sup>2</sup> to correct the measured sample response for the effect of the carbon matrix.

## NONDESTRUCTIVE MEASUREMENTS

### AWCC CALIBRATION

The UO<sub>3</sub> standards used to calibrate the AWCC cover a range of 76 grams to 1523 grams <sup>235</sup>U and are contained in one-liter Nalgene<sup>®</sup> bottles. Each bottle is enclosed in two plastic bags and contained inside a thin walled metal can that is also enclosed inside a plastic bag. Details of the UO<sub>3</sub> standards are presented in Table 1.

TABLE 1. UO<sub>3</sub> standards data

Grams UO <sub>3</sub>	Grams <sup>235</sup> U	$\sigma$ (g <sup>235</sup> U)	Standard Number
100	76.15	0.02	1
300	228.44	0.05	3
600	456.9	0.1	5
1000	761.5	0.2	6
1500	1142.2	0.25	4
2000	1523.0	0.3	2

Two AWCC geometries and two calibration curves were necessary to cover the range of the <sup>235</sup>U quantities in the three material categories. Rag Ash and RCP materials have <sup>235</sup>U contents within the 1523 gram <sup>235</sup>U range of the standards. The AWCC was configured to a 25-cm (10-inch) vertical sample height (with the nickel liner

removed) for these two categories. UO<sub>3</sub> standards were counted one can at a time in this geometry and a calibration curve developed covering the range from 76 to 1523 grams <sup>235</sup>U.

The <sup>235</sup>U content of the PyC-U category falls outside the range of the single can standards. To develop a calibration curve extending to 2284 grams <sup>235</sup>U it was necessary to count two standards together, and this required more internal height in the AWCC counting chamber. Consequently, the geometry of the AWCC was modified by removing two 5-cm (2-inch) poly end plugs provide a 35-cm (14-inch) chamber to accommodate two standard cans stacked vertically. Combinations of two standard cans stacked vertically were counted in the AWCC to produce a calibration curve with a range from 990 grams <sup>235</sup>U to 2284 grams <sup>235</sup>U.

The UO<sub>3</sub> calibration curves are presented in Figure 1. The dashed lines are fitted curves of the form:  $R = \frac{am}{1+bm}$  where R and m are the real coincidence rate (reals rate) in counts per second and the <sup>235</sup>U mass, and a and b are the coefficients of the fit. This formulation represents the data quite well and inverts easily to yield mass as a function of the reals rate R.

### CORRECTION OF THE AWCC RESPONSE FOR THE EFFECT OF CARBON

The LANL scrap materials contain different relative amounts of carbon in addition to their UO<sub>3</sub> content. Carbon and light matrix materials dilute the uranium, decreasing the amount of self shielding, and introducing a slight moderating effect on the interrogating neutron flux. These effects combine to enhance the AWCC response to samples containing carbon relative to the response to non-carbon containing samples. Researchers at LANL have investigated this effect.<sup>1</sup> The data from reference 1 were used to correct for the effect of carbon on the AWCC measurements performed for this characterization. The published data include AWCC calibration curves for a set of U<sub>3</sub>O<sub>8</sub> standards and for a set of standards containing U<sub>3</sub>O<sub>8</sub> in varying amounts of graphite. The

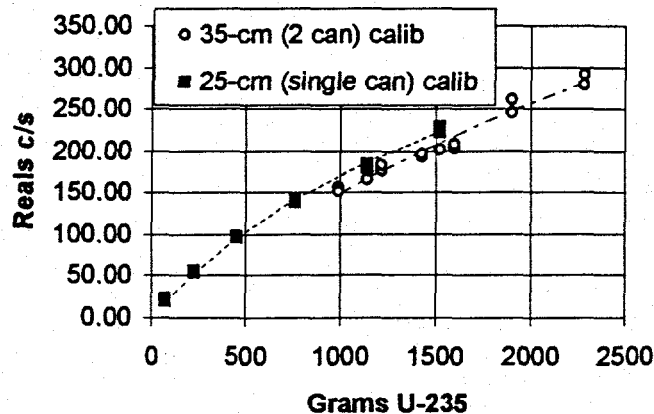


FIGURE 1. Calibration curves for the AWCC in both vertical configurations

published data were fit using standard least squares techniques, and an "enhancement factor" (to account for the increased response due to the carbon matrix) was computed as the ratio of the reals counts per second per gram of  $^{235}\text{U}$  from the carbon-containing curve to those from the non-carbon-containing curve. The calculated enhancement factors were then replotted against the computed C-to-U atom ratios of the carbon-containing standards. The resultant plots were fit to provide a functional relationship between the C-to-U ratio of a sample and the expected enhancement due to the carbon matrix. Figure 2 is the resultant plot.

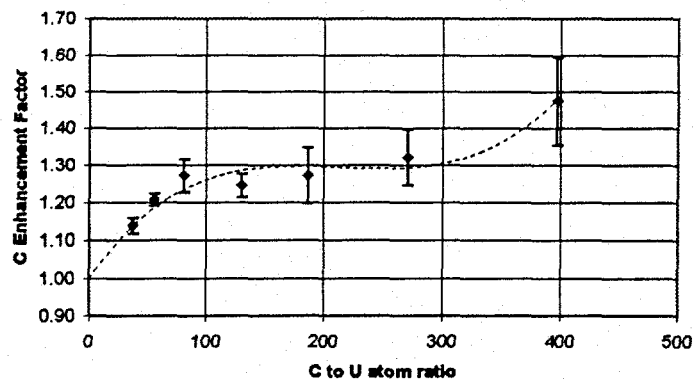


FIGURE 2. Carbon enhancement factor as a function of the carbon-to-uranium ratio<sup>1</sup>

For assay of a carbon-containing sample from the LANL scrap material, the shipper-specified values for sample net weight and weight of uranium were used to estimate the sample's C-to-U ratio. (The samples were presumed to contain only C and U or  $\text{UO}_3$ .) This C-to-U ratio in conjunction with the functional relationship derived from Figure 2 yielded a sample-specific enhancement factor for carbon effect correction. The AWCC reals counting rate measured during assay of each sample was then divided by the computed carbon enhancement factor to correct for the effect of the carbon matrix, and then the corrected counting rate used along with the  $\text{UO}_3$  calibration to compute the  $^{235}\text{U}$  content of the sample.

## RESULTS

Twenty-six samples were analyzed -- 6 RCP, 12 PyC-U, and 8 Rag Ash samples. Each sample, contained in a total of three plastic bags to emulate the standard's packaging, was analyzed in the AWCC and the results corrected for the estimated effect of the carbon content. Counting times were 1000 seconds. The measured  $^{235}\text{U}$  content was converted to total U content using the enrichment specified in the shipping documents. The INEEL's NDA results are compared with the shipping document values in Tables 2 through 4. The quoted uncertainties are one sigma values. In Tables 2-4 the sample result is printed in *italics* if the LANL-specified value is outside of a  $\pm 2\sigma$

TABLE 2. INEEL-measured versus LANL-specified uranium content on selected Rotary Calciner Product (RCP) samples

Sample ID	LANL U (grams)	% $^{235}\text{U}$	C/U ratio	C Enh Factor	INEEL U (grams)
RCX437520B	764	93.15	65.5	1.26	708± 37
<i>RCX347521B</i>	<i>711</i>	<i>93.15</i>	<i>61.6</i>	<i>1.24</i>	<i>586± 30</i>
RCP438616A	186	93.00	242	1.34	192± 17
RCP437550B	322	93.00	133	1.28	301± 19
RCP438619A	373	93.00	138	1.28	370± 23
RCR205010B	109	93.06	143	1.28	126± 11

TABLE 3. INEEL-measured versus LANL-specified uranium content on selected Pyrolytic Carbon-Uranium (PyC-U) Samples

Sample ID	LANL U (grams)	% $^{235}\text{U}$	C/U ratio	C Enh Factor	INEEL U (grams)
EB3506C1	1919	93.15	5.9	1.02	1803± 88
<i>EB3507A1</i>	<i>2038</i>	<i>93.15</i>	<i>5.9</i>	<i>1.02</i>	<i>1852± 91</i>
GAEB3403C2	2087	93.15	5.0	1.02	1967± 98
EB3406C1	1704	93.15	5.0	1.02	1593± 76
EB4000B1	1375	93.15	12.2	1.05	1326± 65
EB3407B1	2331	93.15	5.0	1.02	2150± 110
EB3408A1	2184	93.15	5.0	1.02	2000± 100
EB3503A1	2103	93.15	5.5	1.02	1952± 98
EB3410C1	1551	93.15	5.0	1.02	1481± 73
EB3503B1	2108	93.15	5.9	1.02	1973± 99
<i>GAEB3407A2</i>	<i>2016</i>	<i>93.15</i>	<i>5.0</i>	<i>1.02</i>	<i>1830± 90</i>
EB3404B1	2008	93.15	5.1	1.02	1890± 93

TABLE 4. INEEL-measured versus LANL-specified uranium content on selected Rag Ash Samples

Sample ID	LANL U (grams)	% $^{235}\text{U}$	C/U ratio	C Enh Factor	INEEL U (grams)
ASH006502B	100	93.15	185	1.28	87± 10
<i>ASH454301A</i>	<i>919</i>	<i>45.10</i>	<i>46.1</i>	<i>1.18</i>	<i>1535± 75</i>
ASH300801A	657	93.15	17.3	1.07	688± 34
<i>ASH282901A</i>	<i>959</i>	<i>93.00</i>	<i>30.5</i>	<i>1.12</i>	<i>866± 43</i>
<i>ASH206501A</i>	<i>127</i>	<i>97.64</i>	<i>255</i>	<i>1.35</i>	<i>84± 10</i>
ASH432203B	301	93.15	76.8	1.28	294± 16
<i>ASH431101A</i>	<i>281</i>	<i>34.95</i>	<i>78.5</i>	<i>1.28</i>	<i>356± 30</i>
ASH431102B	97	34.95	89.8	1.28	138± 25

interval constructed about the INEEL AWCC measurement but within  $\pm 3\sigma$ . Results that are outside of a  $\pm 3\sigma$  interval are printed in *underlined italics*.

The INEEL AWCC measurements agreed fairly well with the LANL-specified sample contents; however, the overall method performance was affected by the sample characteristics. The Rag Ash category presented the greatest analytical challenge. The Rag Ash samples are essentially incinerated trash and as such encompass a wide variety of matrix constituents and the widest range of estimated C-to-U ratios. The PyC-U materials have the lowest estimated C-to-U ratios, thus carbon enhancement corrections are small (5% or less.) Of the samples assayed 88% of the LANL-specified values were within a  $3\sigma$  confidence interval constructed about the INEEL measurement, and 73% were within a  $2\sigma$  interval. While these results indicated that our carbon correction procedure had some promise, they could not demonstrate that the method was sufficiently robust for content confirmation on the LANL scrap. Consequently, eight samples selected from the set of 26 analyzed by the AWCC were submitted for destructive analysis.

## DESTRUCTIVE ANALYSES METHODOLOGY

Eight items selected from the LANL scrap materials were destructively analyzed for carbon and uranium content and uranium isotopic ratios.<sup>b</sup> The contents of each container were carefully blended and 5 samples taken from each container for analysis. Uranium assays were via Davies-Gray titrations while the U isotopics were determined by ICP-MS. All standards were NBL certified.

## RESULTS

The chemical analysis results are presented in Table 5. The listed values are the unweighted average of the five analytical results per item. The standard deviations are standard deviations of the average computed from the set of results per item.

TABLE 5. Destructive analysis results for selected items from the LANL scrap inventory

Category	Item Number	% Carbon		% Uranium		U-235 %Enrich	
		Value	$\sigma$	Value	$\sigma$	Value	$\sigma$
RCP	RCP438616A	43.2	3.9	7.24	0.2	93.04	0.03
RCP	RCX437521B	0.052	0	21.42	0.6	93.02	0.01
PyC-U	EB4000B1	43.9	0.7	54.54	0.1	93.17	0.02
PyC-U	EB3407B1	28.8	1.0	66.6	0.5	93.14	0.02
RA	ASH432203B	3.4	1.3	20.8	0.8	89.47	0.15
RA	ASH206501A	4.4	0.9	7.0	0.7	65.3	1.8
RA	ASH 300801A	4.6	0.6	41.3	0.3	90.52	0.16
RA	ASH454301A	4.4	0.1	34.8	0.9	66.7	0.3

Clearly, our previous assumption that these materials are composed entirely of carbon and uranium is incorrect except perhaps for the PyC-U materials. Even if the U is assumed to be in the form of  $UO_3$ , and the extra weight of the oxygen is calculated, for the samples from the RA and RCP categories no more than about half of the sample weight can be accounted for by C, O, and U. Only for the PyC-U materials is the assumption of exclusively C and U reasonably correct.

However, if the unknown matrix materials are light and are not significant neutron poisons, their effect may be very similar to that of graphite.

### DESTRUCTIVE VERSUS NONDESTRUCTIVE COMPARISON

Table 6 compares the destructive and nondestructive assay results. The destructive results for  $^{235}\text{U}$  content per sample were computed in a straightforward manner from the analyses reported in Table 5 and the net weight of each item's contents. The AWCC results are presented both uncorrected and corrected for the assumed "carbon enhancement" effect. The corrections used the shipping documentation data for "C-to-U" ratios.

TABLE 6. Comparison of destructive and nondestructive (AWCC) results for  $^{235}\text{U}$  content

Category	Item Number	Destructive	Uncorrected AWCC		Corrected AWCC	
		U-235 (grams)	U-235 (grams)	% Recovery	U-235 (grams)	% Recovery
RCP	RCP438616A	169.5± 0.3	246± 8	145± 3	179± 16	105 ± 9
RCP	RCX437521B	616± 4	725± 13	118± 2	546± 28	88 ± 5
PyC-U	EB4000B1	1277± 1	1310± 26	103± 2	1235± 61	96 ± 5
PyC-U	EB3407B1	2111± 11	2055± 53	97± 3	1999± 104	95 ± 5
RA	ASH432203B	287± 2	365± 8	127± 2	274± 14	95 ± 5
RA	ASH206501A	82.4± 0.6	112± 5	136± 5	82± 10	99 ± 12
RA	ASH300801A	512± 2	699± 13	137± 2	641± 32	125 ± 5
RA	ASH454301A	755± 7	871± 16	115± 2	692± 34	91 ± 5
AVERAGE % RECOVERY=				122± 17		99.7± 12

The ± values are estimated one-sigma errors. The percent recovery values are the nondestructive (AWCC) result divided by the destructive result expressed as a percentage.

These results show the true value of the carbon enhancement correction procedure. The uncorrected values are biased an average of 22% high while the corrected results average 100% recovery. Only the two items from the PyC-U category, where the correction factor is very small, gave uncorrected recovery values within ± 3 σ of 100%. Only one corrected AWCC result (for ASH300801A) lay outside of a ± 3 σ interval constructed about 100% recovery.

### CONCLUSIONS

These results demonstrate the versatility of the Active Well Coincidence Counter (AWCC.) This work used efficiency normalization factors derived from published AWCC measurements on two different sample forms to correct results obtained on carbon-containing HEU samples at the INEEL. The results showed good agreement with destructive analyses performed on selected samples.

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<sup>b</sup> Chemical analyses were performed by BWX Technologies, Inc., Lynchburg, VA.

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<sup>1</sup> H. O. Menlove, N. Ensslin, and T. E. Sampson, *Experimental Comparison of the Active Well Coincidence Counter with the Random Driver*, LA-7882-MS, June 1979.

<sup>2</sup> H. O. Menlove, *A New Method of Calibration and Normalization for Neutron Detector Families*, LA-11229-MS, April 1988.