

## ENRICHMENT AND URANIUM MASS FROM NMIS FOR HEU METAL

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### ABSTRACT

Recently, the Nuclear Materials Identification System (NMIS) was employed to verify both the mass and enrichment of a number of high-enriched uranium (HEU) metal items stored at the Oak Ridge Y-12 Plant. NMIS was applied for this measurement as an active neutron interrogation system; the measurement performed was similar in nature to a pulsed neutron measurement in that the distribution of detector counts following source emission was accumulated and subjected to analysis.

In order to develop a calibration “surface” versus mass and enrichment, Monte Carlo models of the item geometry were employed to calculate the distribution for a matrix of independently varying masses and enrichments spanning the nominal declared mass and enrichment of the actual items. Each calculated distribution was then decomposed into its moments, and an empirical model of each moment as a function of mass and enrichment was developed. These models were then simultaneously analytically inverted to yield a nonlinear calibration surface that predicted mass and enrichment given the low-order moments of a measured distribution. The uncertainty in the calibration was estimated using standard squared-error minimization and propagation-of-error techniques, and the bias (and its uncertainty) in the calibration was evaluated using measurements of several randomly selected samples.

No single mass or enrichment verified using this method deviated from declaration by more than 5.0%. The typical, i.e., root-mean-squared, error in mass was only 1.6% of the declaration, and the typical error in enrichment was only 1.5% of the declaration. As a result, a finding previously issued for this storage area by the Department of Energy, Oak Ridge Operations, (DOE-ORO) was resolved.

### INTRODUCTION

In 1999, DOE-ORO issued a finding for a particular storage area at the Oak Ridge Y-12 Plant; to be resolved, this finding required that the mass and enrichment of a number of HEU metal items stored in the area be verified. However, the Oak Ridge Y-12 Plant Nuclear Materials Control and Accountability (NMC&A) Organization determined that it could not, by standard techniques, *safely* verify the mass and enrichment of HEU metal items in this particular storage area due to the significant potential for the spread of airborne contamination. Ordinarily, the mass of these items would have been measured using a scale, and their enrichment would have been estimated using passive gamma spectrometry. However, the former required the items to be removed from their storage containers, and the latter was similarly difficult to implement unless the items were removed from their storage containers. Unfortunately, no hood that could be transported to the storage area was available, and radiological control dictated that a fixed hood outside the area could not be used extensively without risking the spread of airborne contamination to the rest of the facility. In any event, the only suitable fixed hood in the same facility was at that time dedicated to higher priority tasks.

Consequently, the Oak Ridge National Laboratory was asked if it would be possible to use the Nuclear Materials Identification System (NMIS) to verify the mass and enrichment of these items in-situ, i.e., without removing them from their containers and without removing the containers from the storage area. Chiang, et al., describes the operational aspects of these verification measurements.<sup>1</sup>

Prior to this, NMIS had never been used to estimate the enrichment of uranium metal. After performing a series of measurements on ten samples selected at random from the storage area, the subsequently described method to simultaneously estimate both the mass and enrichment of these HEU metal items was developed.

## METHOD

NMIS was operated as an active neutron interrogation system for this application. A small ( $\sim 1 \mu\text{g}$ )  $^{252}\text{Cf}$  ionization chamber was employed as the active source and an array of fast plastic scintillators was used to detect neutrons and gammas emitted by the inspected items in response to the source. NMIS acquires a variety of statistics, but early in this application it was determined that only the distribution of detector counts following  $^{252}\text{Cf}$  fission would be subjected to analysis in order to estimate the mass and enrichment of the inspected items. In this respect, the measurement was similar in nature to a pulsed neutron measurement.

The items to be inspected exhibited only small deviations from nominal in the declared mass and enrichment. So, in order to develop a calibration surface versus mass and enrichment, MCNP-DSP models of the item geometry were employed to calculate the distribution for a matrix of independently varying masses and enrichments.<sup>2</sup> The masses and enrichments modeled were selected to span the nominal declared mass and enrichment of the actual items.

Each calculated distribution  $R(\tau)$  of detector counts following  $^{252}\text{Cf}$  fission, where  $\tau$  denotes the time-delay between  $^{252}\text{Cf}$  fission and detection events, was then decomposed into moments of the form

$$M_n = \sum_k \tau_k^n R(\tau_k) .$$

In particular, note that the zeroth moment  $M_0$  is simply the source-correlated detector count rate (i.e., the area under the distribution) and the first moment  $M_1$  is the product of the source-correlated count rate with the distribution-weighted mean delay  $\bar{\tau}$ :

$$M_1 = M_0 \cdot \bar{\tau} = M_0 \cdot \frac{\sum_k \tau_k R(\tau_k)}{\sum_k R(\tau_k)} .$$

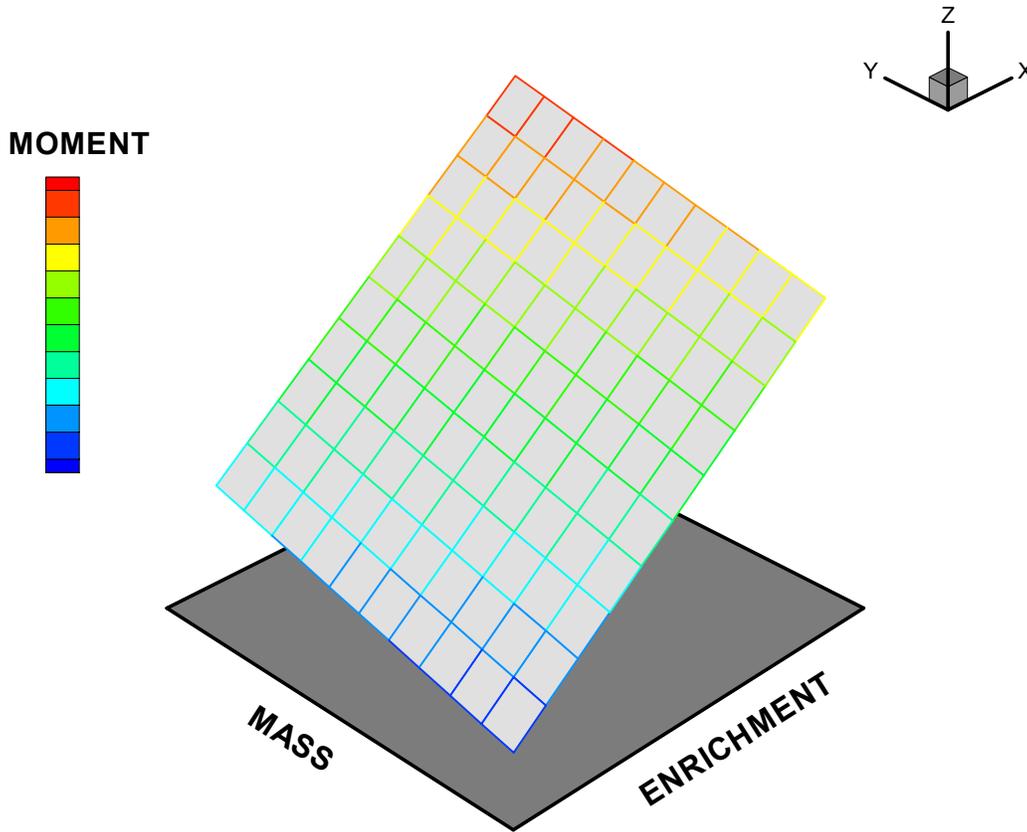
Higher order moments similarly yield the product of the source-correlated count rate with the mean-squared delay, mean-cubed delay, etc.<sup>a</sup>

Figure 1 illustrates the decomposition of the distribution into moments over the matrix of independently varying masses and enrichments; this decomposition results in a surface that yields a particular moment at a given (mass, enrichment) pair. Empirical models predicting these moments given the mass and enrichment were developed and the model parameters (and their uncertainty) were estimated using standard squared-error minimization techniques. To develop these models, the dependence of each moment as a function of only mass for a fixed enrichment was determined. This dependence was found to have the same trend for all moments and for all enrichments; only the parameters of each trend varied with enrichment. The dependence on only enrichment of these parameters was subsequently determined

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<sup>a</sup> Recall that a distribution can be reconstructed from its moments exactly if all the moments are known (A. Papoulis, *Probability, Random Variables, and Stochastic Processes*, 2<sup>nd</sup> Ed., McGraw-Hill, 1984). In other words, the moments are just an alternate representation of the distribution. Hence, using a few low-order moments to represent the distribution is essentially a rudimentary compression technique.

to produce a family of models that predict a given moment as a simultaneous function of mass and enrichment.



**Figure 1. Illustration of a surface representing a decomposition moment over a matrix of independently varying masses and enrichments**

These models were then simultaneously analytically inverted to obtain a nonlinear calibration surface of the form

$$\begin{aligned} \text{mass} &= f(M_0, M_1) \\ \text{enrichment} &= g(M_0, M_1) \end{aligned} \quad ,$$

that predicted mass and enrichment given the two lowest order moments ( $M_0$  and  $M_1$ ) of a measured distribution. The uncertainty in the calibration was estimated from that of the model parameters using standard propagation-of-error methods.

The Monte-Carlo-calculated calibration was not without error, i.e., it yielded a bias in the estimated mass and enrichment when applied to a measured distribution. This bias, and its uncertainty, was evaluated by measuring ten samples randomly selected from the inventory of items to be inspected. These ten items were set aside for independent verification using a scale and gamma spectrometry. It must be noted that *only* these ten items were independently verified. The preceding model, once corrected for the bias in the Monte Carlo calculations, was then applied to verify the mass and enrichment of *all* other inspected items.

## RESULTS

No single verification using the preceding method yielded an estimated mass or enrichment that deviated from the declared by more than 5.0% (see Fig. 2 for typical results and Table 1 for a summary of the results). The root-mean-squared (RMS) deviation of the estimated mass relative to the declared mass was only 1.6%, and the RMS deviation of the estimated enrichment relative to the declared enrichment was only 1.5%. The (arithmetic) mean deviation of the estimated mass relative to the declared was +0.2%, and the mean deviation of the estimated enrichment relative to the declared was  $-0.2\%$ . The RMS relative uncertainty of the mass estimate was 5.3% of the estimate, and the RMS relative uncertainty in the enrichment estimate was 9.6% of the estimate.

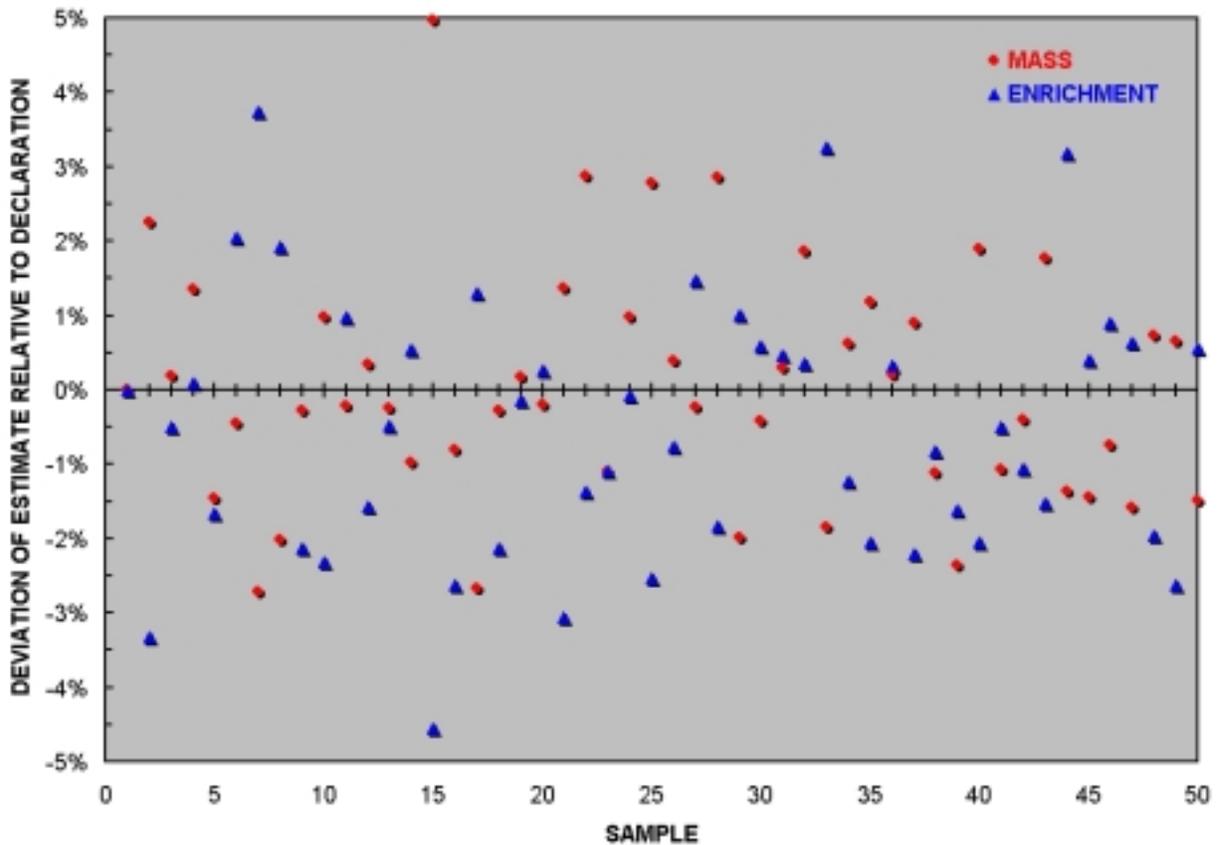


Figure 2. Typical relative deviations in the verification of mass and enrichment

**Table 1. Summary of relative uncertainty and deviations in the verification of mass and enrichment**

		Mass	Enrichment
<b>RMS Relative Uncertainty</b>		5.3%	9.6%
<b>Relative Deviation</b>	<b>Minimum</b>	-4.0%	-4.6%
	<b>Maximum</b>	5.0%	3.7%
	<b>RMS</b>	1.6%	1.5%
	<b>Mean</b>	0.2%	-0.2%

It is important to note that this method, as it currently exists, is probably *not* generally applicable to all uranium metal items. The parameters of the calibration surface are unlikely to be strictly independent of geometry and are certainly applicable only over a limited range of masses and enrichments. However, it *is* likely that with further development, which would necessarily include reconfiguration of the source and detectors to minimize time-of-flight and other geometry-related effects, this method could be extended to a wide variety of uranium metal items.

## CONCLUSIONS

These NMIS measurements were completed expeditiously; each verification took only 6 minutes including the time required to change the sample. Furthermore, no inspected item was removed from its container and no inspected container was removed from the storage area. No spread of airborne contamination was detectable.

Following the completion of the verifications and the submission of the results, NMC&A and DOE-ORO accepted the declared mass and enrichment of each item and thereby resolved the finding. The items were subsequently sealed and now only periodic inspection of randomly selected samples is required.

## REFERENCES

1. L. G. Chiang, J. K. Mattingly, J. A. Ramsey, and J. T. Mihalcz, "Verification of Uranium Mass and Enrichment of Highly Enriched Uranium (HEU) Using the Nuclear Materials Identification System (NMIS)," Proceedings of the Institute of Nuclear Materials Management 41<sup>st</sup> Annual Meeting, July 2000.
2. T. E. Valentine and J. T. Mihalcz, "MCNP-DSP: A Neutron and Gamma Ray Monte Carlo Calculation of Source Drive Noise-Measured Parameters," *Ann. Nucl. Energy* **24**, 1271, 1996.