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High Precision Nondestructive Assay to Complement DA

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Abstract

Large scale spent fuel reprocessing plants and fuel fabrication facilities require large numbers of grab samples taken for destructive analysis (DA) for nuclear material accountancy and safeguards. The resulting cost and radioactive waste generation is large and significant time is lost between the sample taking and the DA results. This paper presents an approach to combine nondestructive assay (NDA) with DA calibration and normalization to reduce the number of chemical analysis and shorten the time to obtain the results. For plutonium bearing samples, the preferred NDA system is a modified version of the high efficiency epithermal neutron counter. These are well-type neutron counters with efficiencies in the range of 60-70% and short neutron die-away times. The counting precisions of the new counters are an order of magnitude better than for prior thermal-neutron counters for multiplicity analysis. The accuracy goal for the NDA is in the range of 0.2-0.5% for the Pu-240 effective mass. The stability and statistical precision of the neutron counters are better than 0.1%, and the total measurement error will be dominated by how well the standards from the DA laboratory match the samples measured with the NDA. The analysis of the neutron data can be performed in the simple coincidence (doubles) mode, or the "known-alpha" mode, or the "multiplicity" mode. Each of these modes has advantages and disadvantages depending on the experimental conditions. This paper will evaluate the analysis methods to obtain to obtain the best accuracy. Also, the preliminary design of a neutron counter that is optimized for the measurement of small samples such as powder and pellets will be presented.

Introduction

The implementation of large scale reprocessing plants and MOX fabrication facilities during the past two decades has led to a large increase in the number of chemical destructive analysis (DA) of small inventory samples. The analysis of these samples is required for material control and nuclear safeguards; however, the DA procedure is time consuming, costly, and significant amounts of nuclear wastes are generated. The purpose of this paper is to present an option of using nondestructive assay (NDA) to supplement the DA and thereby reducing the DA throughput without sacrificing the inventory accuracy. The cost and timeliness of the results can be significantly improved with the combination of NDA with the DA.

Prior work for the EURATOM On-Site Laboratories [1] has demonstrated inventory sample neutron coincidence rates for small powder samples with statistical uncertainty of 0.2% with measurement times of 2 to 30 hours. The total uncertainty was greater than this value because of the error associated with the HRGS measurements of the plutonium

isotopic ratios. For the present application, we plan to use the mass spectroscopic values for the Pu ratios and use HRGS for confirmatory measurements.

During the past decade, there have significant improvements in neutron counters that use He-3 tubes to measure the neutrons emitted from the Pu-240 effective in the inventory samples. The epithermal-neutron multiplicity counters (ENMC) [2] use higher-pressure He-3 gas mixtures combined with reduced thickness of high density polyethylene to increase the efficiency and to shorten the neutron die-away time. This combination of pressure and moderator changes result in an improvement in the statistical precision of a factor of 5 to 20 compared with traditional thermal-neutron counters depending on the characteristics of the samples. Asano et al. [3] have demonstrated statistical errors for small samples of less than 0.15% using an ENMC with a 100 minute measurement time.

A number of ENMC type systems have been specifically designed for inventory sample measurements. These counters differ from the standard ENMC in that the sample cavity is smaller. The efficiency is higher and the die-away time is shorter than standard inventory sample counters. This paper will describe the analysis methods, the detector system, the calibration procedures, and the design performance.

Measurement Concept

The inventory sample measurements are intended to supplement the DA and not replace it. The neutron measurement provides the Pu-240 effective and the mass spectrometry isotopic ratios are used to get the total plutonium mass in the samples. The DA standards are used to calibrate the NDA system and to perform quality control and bias checks. For example, the NDA system would measure 100% of the sample throughput and the DA would measure 10% of the same sample set. The absolute calibration of the NDA system comes from the DA standards and any systematic bias between the NDA and the DA would be corrected by adjusting the calibration parameters in the NDA system. The primary function of the NDA is to reduce the DA load, but the calibration of the NDA comes from the DA.

For optimum implementation, the NDA needs to have a precision and stability that is as good as the DA target of approximately 0.2% for the Pu-240 effective. The primary sources of error for the ENMC small sample measurements are the calibration standards, statistical precision, sample positioning, and system counting stability. The uncertainty in the Pu isotopic ratios is common for the DA and NDA and is not part of the NDA error estimate for the Pu-240 effective. The error will be smaller than would be the case for an absolute Pu measurement because we are only measuring the Pu-240eff and the batch isotopic ratios for Pu are the same as determined via DA.

The statistical counting error depends on the counting time and system stability. The stability has been shown to be better than 0.01% [4]. Neutron multiplicity counters provide singles, doubles, and triples (three neutrons detected from the same fission reaction) counting rates. The doubles and triples backgrounds are usually very small and can be measured for long periods; however, the singles background needs to be measured

near to the time of the sample measurement to correspond to potential nuclear materials in the room. The inventory samples should provide a counting rate that is substantially higher than the background.

The calibration of the NDA system is performed using DA standards of each material category of interest such as Pu powder, MOX powder, MOX pellets, and liquids. Thus, any difference between the material types is taken into account by calibrations using the same materials. Also, the sample containers and positioning device are the same for the calibration standards and the unknowns. These steps are necessary to keep the associated errors below ~ 0.15%.

Three Calibration and Measurement Options

The measurement of the inventory samples can be evaluated using three different methods:

a) Measured doubles versus Pu-240e mass ("passive calibration curve")
For measurement situations where the singles neutron background has a significant uncertainty, the most accurate results might be obtained using the uncorrected doubles rate. This is especially true for small samples where the neutron multiplication is small. For this mode of calibration, a well matched set of standards is required and the multiplication is built into the calibration curve.

b) "Known-Alpha" multiplication correction
The Known-Alpha calibration mode can be used when the material is pure so that the alpha factor is accurately predicted by the isotopic ratios. This mode of measurement always has the best statistical precision and the calibration is a straight line. If there is an error related to sample purity the resulting assay values are always high.

c) Multiplicity Mode
This mode of assay will be our primary focus for the ENMC type instruments because both the purity and the multiplication can be different from the standards. The ENMC instruments have a high efficiency and short die-away time to make the statistical precision meet our 0.2% requirements. However, the singles background rate must be accurately measured, and shielding on the outside of the neutron detector is used to reduce the singles neutron background. The room background for doubles and triples is very small and relatively constant.

In practice, the INCC software permits the analysis with all three methods for the same data set. A comparison of the answers then can be used to identify possible inaccuracies in the singles background subtraction.

ENMC Description
Figure 1 shows cross sections through Monte Carlo Models of a traditional inventory sample counter (INVS) [5] and an ENMC designed for small samples. The large increase in the number of tubes and the reduction in spacing can be seen. The ^{3}He pressure in the

ENMC case is 10 atmospheres compared to 4 atmospheres in the INVS. These differences lead to a system with much higher detection efficiency and lower dieaway time. The ENMC uses standard Amptek A-111 amplifiers that have been used in neutron coincidence counters since the 1980s. The counting rate for small samples is low and the errors of deadtime do not contribute significantly to the overall uncertainty. The total signal from all detectors is used for the mass determination. In addition the signals from the separate rings of detectors are available. The counting rate ratio between the inner and outer rings is sensitive to the neutron energy emitted from the sample and can be used as an indicator of impurities in the sample.

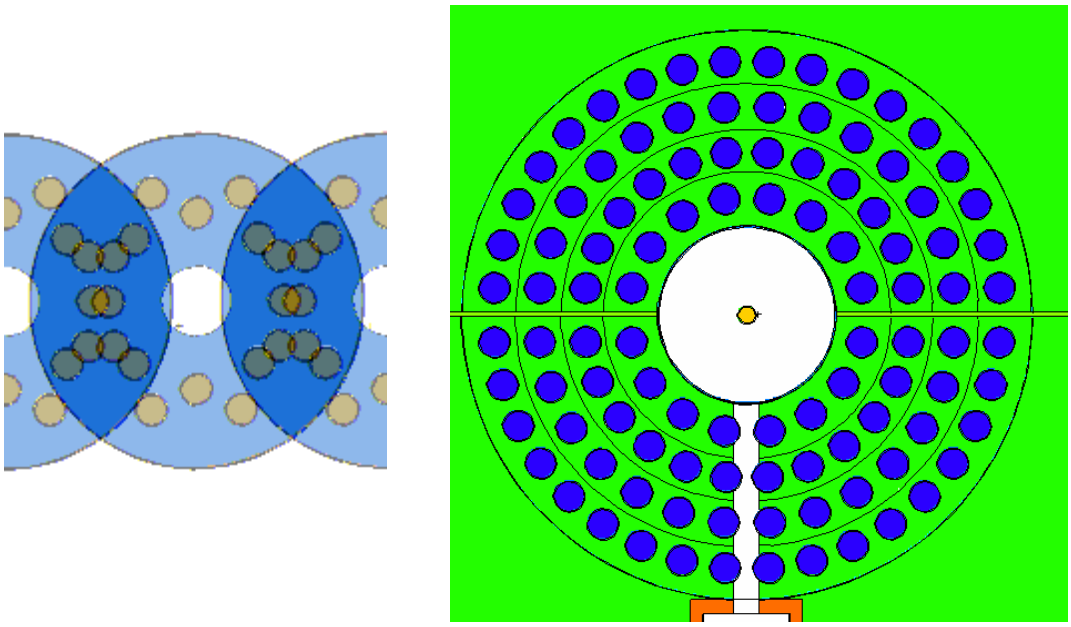

Figure 1 Horizontal cross-sections of Monte Carlo models of an INVS (left) and an ENMC(right)

Physics Parameters

The ENMC detector shown in figure 1 has a neutron detection efficiency for a point ^{252}Cf source of 62.8%. For a point ^{240}Pu source the efficiency is 63.8% because of the softer neutron spectrum. The neutron dieaway time of the system is 20 μs. The detector has a doubles gate fraction of 0.647 and a triples gate fraction of 0.418 when a predelay of 1.5 μs and a 24 μs gate are used.

Performance Estimates

a) Random Errors

Table 1 shows the results of a simulated one hour measurement of a small sample containing 4.4 g of plutonium in an ENMC.

Table 1 Uncertainty from Counting Statistics on small Pu mass measurement (1 hour)							
	Singles Cps	Doubles cps	Multiplication Corrected Doubles cps	Triples cps	Passive Calibration Curve Mass (g)	Known alpha mass (g)	Multiplicity Mass (g)
Mean value	2084.9	414.2	404.2	82.31	4.39	4.28	4.27
Rel error %	0.06%	0.14%	0.07%	0.41%	0.14%	0.07%	0.12%

The table shows the statistical uncertainty on the raw singles, doubles and triples counting rates, which are 0.06%, 0.14% and 0.41% respectively. It also shows that the statistical error on the multiplication corrected doubles counting rate is 0.07%. The values given for the calculated Pu mass are not important, but the resulting errors on the masses show the optimum performance of a 1 hour measurement. The passive calibration curve method gives an uncertainty of 0.14%, the known-alpha method gives an error of 0.07% and the multiplicity method gives an uncertainty of 0.12%. It should be noted that these estimates do not include any contribution from the errors on the background. Such errors would affect the known-alpha and multiplicity methods; the detector will be well shielded to avoid a significant contribution from background.

b) Systematic Errors

Potential systematic errors include the system stability, the counting statistics from the calibration experiment and any difference between the calibration standard and the measurement of the unknown. The latter includes such things as positioning, container, moisture, and density. It is assumed that there will be a specific calibration standard for each material type and each container type. The system stability is typically better than 0.01%, as given above. The counting statistics on the calibration experiment can be taken from the previous section. For this type of sample, a sample holder can be used to give very reproducible positioning (<1 mm) in the sample cavity. In this case, the distribution of material in the container has more influence on the uncertainty than the container position. Asano et al. [3] estimated this contribution to be 0.1% for small samples. The remaining factors are moisture (or other impurities) and density. Table 2 shows the results of simulated measurements for two cases. In the first case 5% by weight of moisture was added to the sample and in the second case the sample density was increased by 10%. The magnitudes of these values were chosen to see the sensitivity of the different methods to such changes rather than to represent the values expected during real measurements. Moisture affects the neutron spectrum in the sample and hence the multiplication and efficiency, as well as increasing the (α,n) production. (The (α,n) production is usually given in terms of the "alpha" value, which is equal to the (α,n) production rate divided by the spontaneous fission neutron production rate). Table 2 shows that the singles rate of the sample increases by about 9%. The doubles rate is much less affected and only increases by 0.72%. The overall effect is to change the passive calibration mass by 0.59%, the known-alpha mass by 3.23% and the multiplicity mass by

0.29%. The small change in multiplicity mass from such a large change in moisture content shows how insensitive the method is to moisture. The multiplicity method measures an "alpha" value of 1.044 compared to the value of 1.039 that was used in the simulation. The "known-alpha" value for alpha is 0.867.

Table 2 Uncertainty from Counting Statistics on small Pu mass measurement (1 hour)

	Singles Cps	Doubles cps	Multiplication Corrected Doubles cps	Triples cps	Passive Calibration Curve Mass (g)	Known alpha mass (g)	Multiplicity Mass (g)
Change with 5% moisture	9.25%	0.72%	3.23%	1.33%	0.59%	3.23%	0.29%
Change with 10% higher density	0.23%	1.45%	-0.02%	4.5%	1.44%	-0.02%	-0.20%

The simulated measurement with the powder density increased by 10% shows the expected pattern. The passive calibration curve mass result increases by 1.4% as the multiplication in the sample increases. The known-alpha result remains essentially unchanged as the correction method removes the effect of multiplication. The multiplicity result changes by 0.2%. The difference between "pour" and "tap" density of PuO2 powder is in the range of 20-40% [6]. This implies that by treating all of the powder samples in the same way, it should be possible to reduce the density variations to much less than the 10% value used in these results. This means that the contribution from density changes would be of the order of 0.7%, 0% and 0.1% for the passive calibration curve, known-alpha and multiplicity methods respectively.

Summary

This paper has outlined a measurement concept for non-destructive analysis of small samples in support of destructive analysis. NDA measurements give advantages in terms of the elapsed time to obtain results, reduced waste and reduced overall cost. The concept involves careful calibration with representative standards for all material types of interest and the use of isotopic composition from destructive analysis. In this way many of the systematic errors associated with neutron counting do not contribute to the final uncertainty. Three analysis methods can be used: passive calibration curve, known-alpha and multiplicity analysis. These three methods are affected differently by impurities and density and have different precision. Comparing the results gives more information on the sample and the reliability of the result. Values have been given for the contributions to the uncertainty expected for neutron multiplicity measurements of small samples. The conclusion is that an uncertainty of around 0.2-0.5% is a realistic target.

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