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ASSAY OF IMPURE PLUTONIUM OXIDE WITH THE LARGE NEUTRON MULTIPLICITY COUNTER FOR IAEA VERIFICATION OF EXCESS WEAPONS MATERIAL AT THE ROCKY FLATS ENVIRONMENTAL TECHNOLOGY SITE

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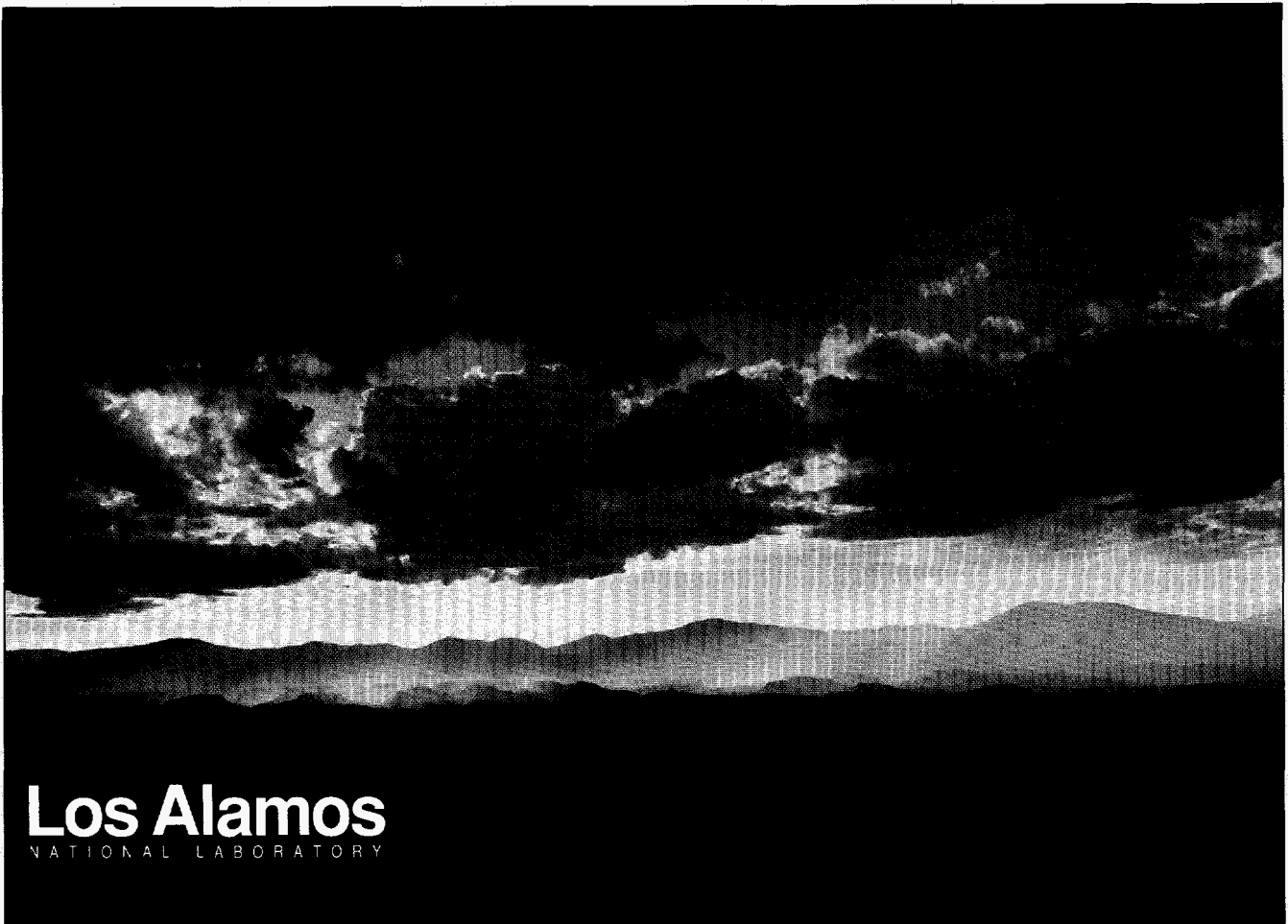
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**Assay of Impure Plutonium Oxide with the Large Neutron Multiplicity Counter for
IAEA Verification of Excess Weapons Material at the
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Abstract

The large neutron multiplicity counter (LNMC), also known as the 30-gal.-drum neutron multiplicity counter, has now been used successfully by the International Atomic Energy Agency (IAEA) for the Initial Physical Inventory Verification (IPIV) and the first annual Physical Inventory Verification (PIV) of excess weapons plutonium oxide at the Rocky Flats Environmental Technology Site (RFETS). These excess plutonium oxide materials contain a variety of impurities. They are stored two cans to a 10-gal.-drum. The drums contain from 1.3 to 4.0 kg of plutonium. The isotopic declarations vary from can to can but the material averages 6% ^{240}Pu .

During the IPIV, 94 samples were measured in the LNMC; 19 were measured during the PIV. The assays for all but a single drum agreed to within three standard deviations of the declared value. This problematic drum could not be measured by the LNMC because of its unusually high neutron emission rate. In this paper we will report on the overall performance of the LNMC in these inspections.

Introduction

Inspections of excess weapons plutonium at Rocky Flats Environmental Technology Site (RFETS) by the International Atomic Energy Agency (IAEA) are now in their second year. During this period, the inventory of approximately one metric ton of pure and impure plutonium oxide materials has been sampled and measured using neutron multiplicity counting and destructive analysis. The database of measurements now includes measurements of over 100 drums made with the large neutron multiplicity counter (LNMC) and destructive analysis of samples from 50 cans. The materials at RFETS are stored in 10-gal. drums as shown in Fig. 1. Nominally 2 kg of plutonium oxide is contained in each can centered in the drum by a steel tube or spider. About one-third of the drums also have lead liners to reduce personnel radiation exposure; however, this has no effect on the neutron measurements.

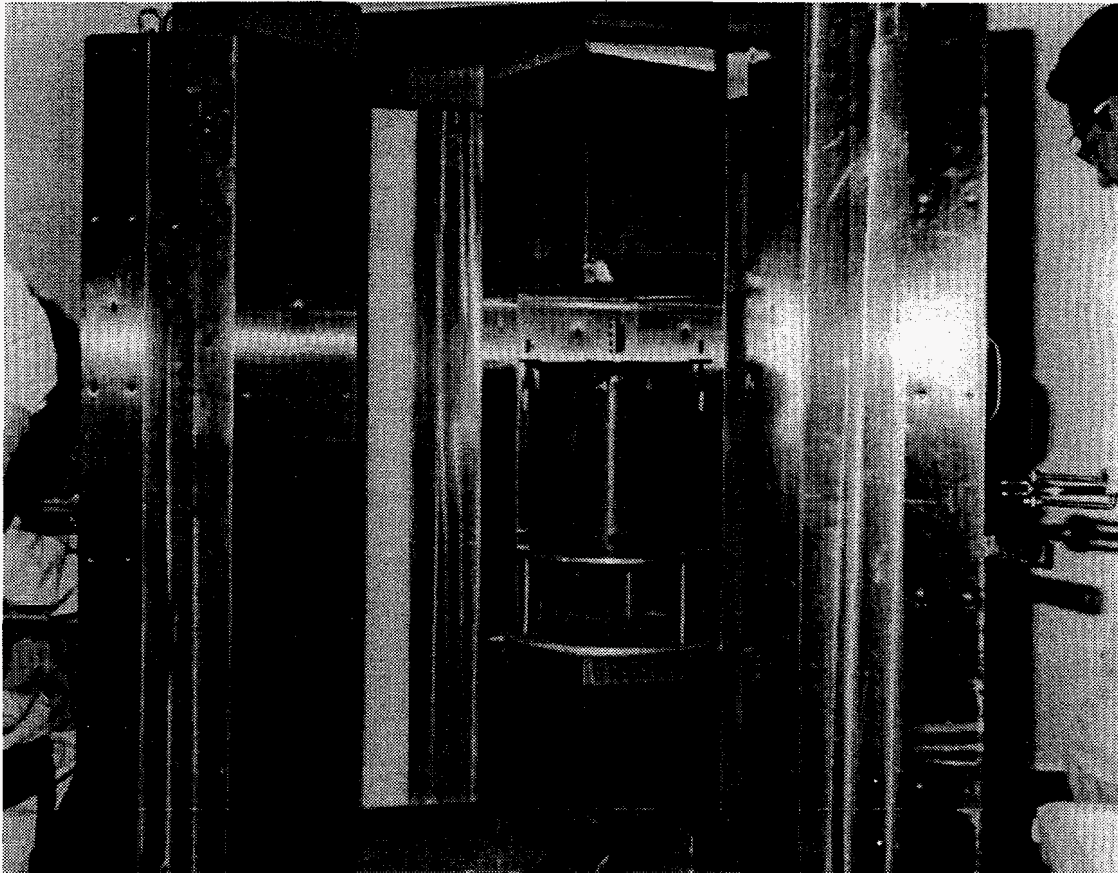


Fig. 1. A 10-gal. drum sample in security overpack in the LNMC during the PIV at RFETS.

The LNMC, shown in Fig. 2 during use at RFETS, was originally designed¹ to measure weapons components stored in 30-gal. drums. Its design criteria were that it provide assays to a precision of from 1% to 3% in 30 minutes for samples whose mass ranged from 2 to 5 kg of plutonium and whose impurity levels were such that at least half of their total neutron emissions were from spontaneous fission. Initial testing and calibration of this instrument and its identical twin, now at the Lawrence Livermore National Laboratory (LLNL), demonstrated that the instrument met or exceeded these criteria.² The materials at RFETS meet the first criteria; however, more than 80% of the oxide materials have impurity levels higher than the design criteria. Thus it was anticipated that the overall precision would be slightly worse than 3% for these samples.

As was reported last year,³ the neutron multiplicity results obtained with the LNMC during the Initial Physical Inventory Verification (IPIV) were in excellent agreement with operator-declared values. The operator-declared values are derived from calorimetric and gamma-ray isotopic measurements analyzed by GRPAUT⁴ or its TRIFID counterpart, EPICS,⁵ between 1985 and 1990. However, at that reporting the destructive analyses were not yet completed and thus no conclusions could be drawn concerning the counter's performance relative to destructive analysis. Since then the first annual Physical Inventory Verification (PIV) has been completed, and destructive analysis results have been received.

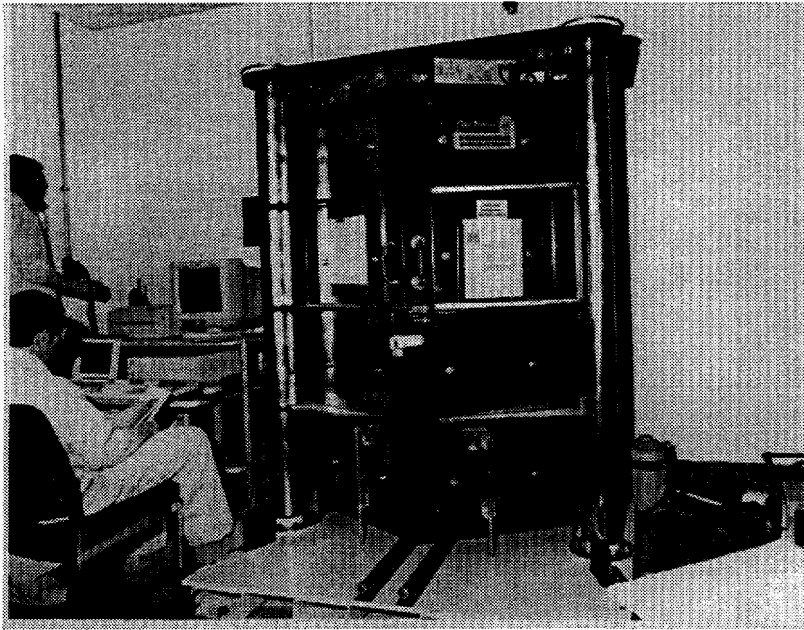


Fig. 2. The large neutron multiplicity counter at RFETS.

LNMC Performance Relative to Site Declarations

Figures 3 and 4 and Table I give the overall performance of the LNMC relative to operator-declared values. In all cases, the LNMC yielded assays that agreed within three standard deviations of the declared values. The average deviation is within two standard deviations of zero. The combined data suggest that a small bias may exist. However there is only a 33% probability that this bias is real, assuming normal statistics. For all samples the average assay was within 4.2% of the operator declaration 63% of the time.

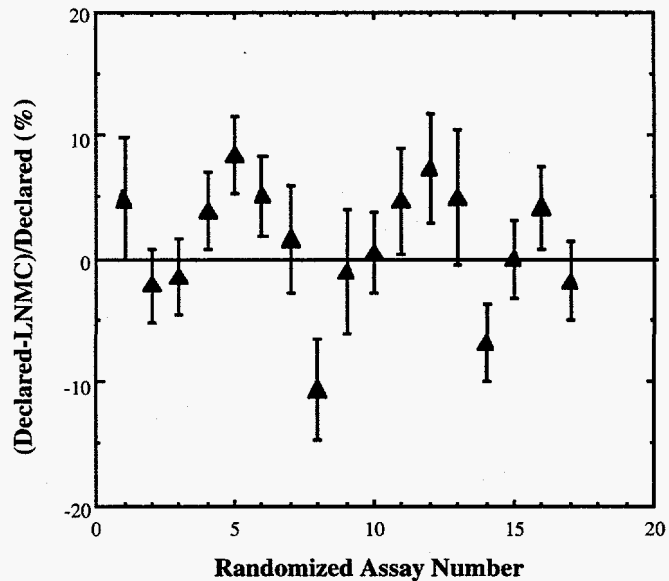


Fig. 3. Multiplicity assay results obtained with the LNMC during the PIV at RFETS.

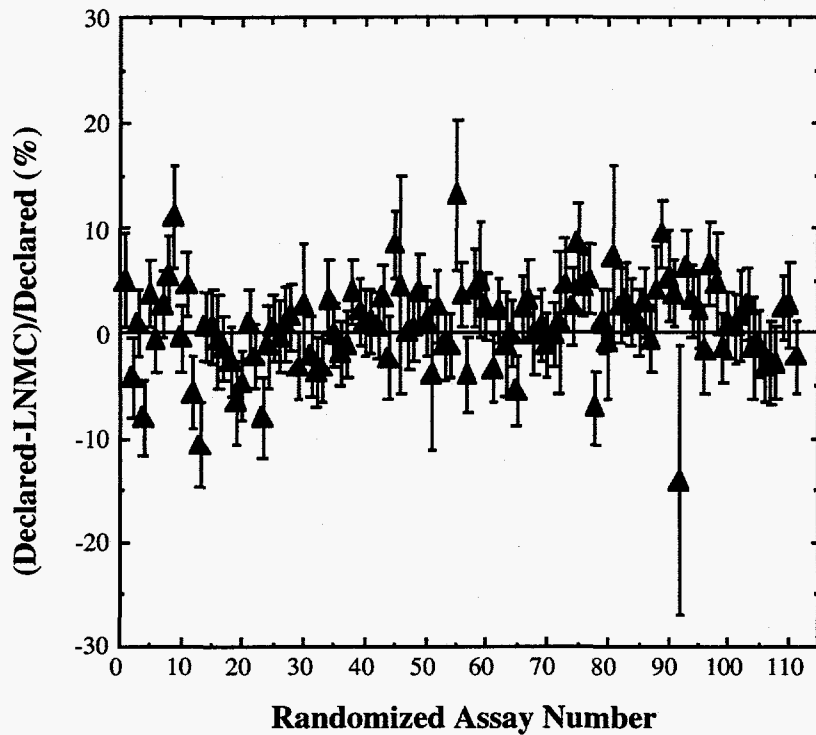


Fig 4. Combined multiplicity assay results obtained with the LNMC during the IPIV and PIV at RFETS.

Table I. LNMC Results for Verifications			
	IPIV	PIV	All
Number of Measurements (N)	93	18	111
Average [Declared (D) - Assay (A)] / Assay (%)	0.61	1.28	0.75
1 sigma	4.11	4.99	4.17
1 sigma / \sqrt{N}	0.42	1.18	0.40

LNMC Performance Relative to Destructive Analysis

Twenty-five of the drums in the inventory were sampled for destructive analysis. Of these, 24 of the drums were successfully measured with the LNMC. The only drum that failed to be assayed successfully by the LNMC had a measured neutron rate in excess of 2.7×10^6 counts/s, a rate too high for the multiplicity shift register to process without data loss. This sample will be discussed below.

Of the 24 drums measured in the LNMC, 16 contained cans that had homogeneous characteristics as evidenced by the agreement between the destructive analysis of the individual samples taken from the cans. The other eight drums had one can that displayed heterogeneous characteristics.

Figure 5 and Table II show the performance of the LNMC relative to the destructive analysis results. The table also includes the agreement with operator declarations for comparison. Note that

the agreement between the LNMC and destructive analysis results is worse, although not dramatically so, for the drums containing heterogeneous cans than the agreement between the LNMC and operator declarations. This is most likely the result of sampling error for destructive analysis. Both the neutron multiplicity technique and the calorimetric technique used to obtain the operator declarations sample the cans completely; whereas the destructive analysis must rely on discrete samples. When an item is heterogeneous, the true nature of the material cannot be sampled unless the material is pulverized and blended. This was not done for the RFETS materials.

However, the neutron multiplicity results are also generally worse for the heterogeneous samples compared to the homogeneous ones. This is because the heterogeneous items tended to contain more low atomic number impurities than the homogeneous items and these impurities tend to

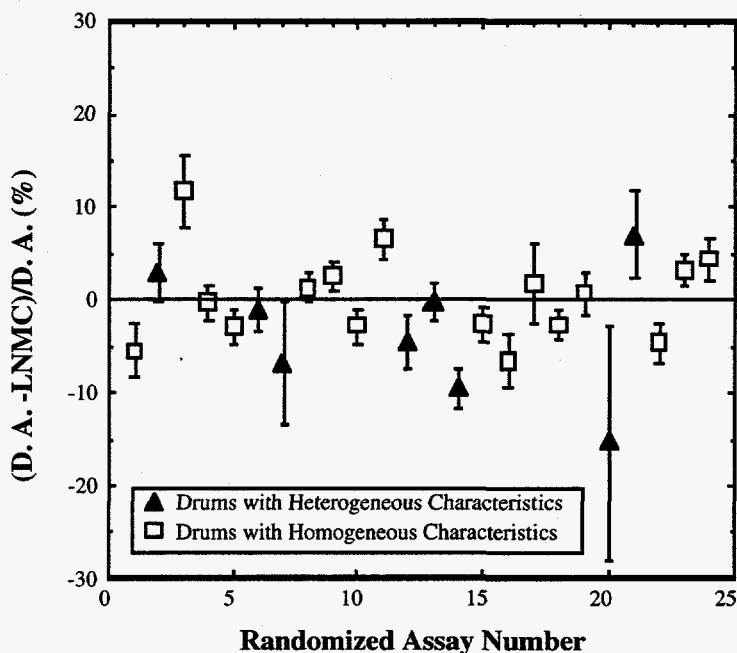


Fig. 5. Multiplicity assay results obtained with the LNMC compared with destructive analysis results for 24 drums.

Table II. LNMC Compared to Destructive Analysis and to Operator Declarations for 24 Drums Sampled for Destructive Analysis (DA)						
	(Operator - LNMC)/ Operator (%)			(DA - LNMC) / DA (%)		
	Average	1 sigma	1 sigma of mean	Average	1 sigma	1 sigma of mean
All Samples	-0.79	5.24	1.07	-0.97	5.79	1.18
Drums with Homogeneous Characteristics	-0.43	4.90	1.22	0.26	4.79	1.20
Drums with Heterogeneous Characteristics	-1.49	6.15	2.18	-3.43	7.12	2.52

decrease the precision of the multiplicity assay. Figure 6 shows the measurement precision due to counting statistics relative to the ratio, α , of spontaneous fission emissions to (α, n) emissions arising from low atomic number impurities. From this plot, there is a higher fraction of high α samples in the heterogeneous population than the homogeneous population sampled. Thus the average precision is poorer for the heterogeneous population, and one would expect the average agreement to also be poorer.

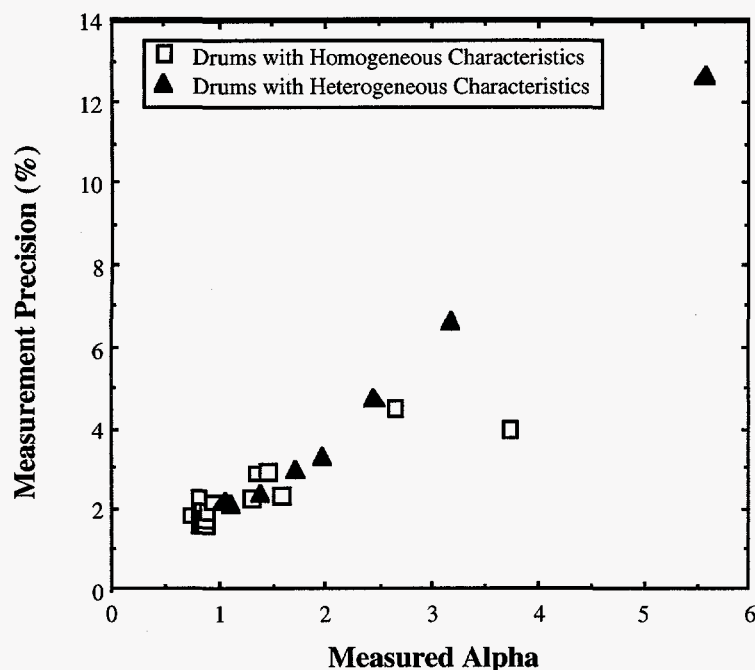


Fig. 6. Multiplicity assay precision due to counting statistics vs the ratio, α , of (α, n) neutrons emitted to spontaneous fission neutron.

Drum with High Neutron Emissions

As mentioned above, there was one drum selected for both the IPIV and the PIV that the LNMC could not measure because the neutron rate detected exceeded what the multiplicity shift register could process. At the IPIV the decision was made to sample the drum for destructive analysis. During this analysis, one of the cans in the drum was found to be heterogeneous; although overall the destructive analysis agreed well with the operator declarations. When the drum was selected again for measurement during the PIV, we decided to experiment with the LNMC to see if we could measure the sample by reducing the efficiency of the detector by turning off one or several of the detector banks. Unfortunately with only one bank turned off, the multiplicity distribution was too long for the deadtime correction algorithms to process reliably. So we turned off a second bank. With two banks turned off, the LNMC's detection efficiency is reduced from 42% to 28%. This reduces the ability to detect triples by more than a factor of 3.

Because the accidental rate was clearly very high for this sample, there was little hope of obtaining sufficient statistical precision in the triples to do an assay in any reasonable period of time. An assay based on a 17-h count still gave anomalous results. If we assume that the operator declaration

for this drum is correct and we assume that the sample self-multiplication is approximately the same as samples in this population having similar mass, the neutron data obtained with only four of the six banks turned on suggests that alpha for this drum is in excess of 40.

The ratio of counts in the inner row of the counter to the outer row gives some information about the impurity causing this high neutron rate. Figure 7 shows this ratio for all the drums measured during the IPIV and the PIV. The sample with the very low ratio is the drum with the high rate. This suggests that the impurity in this drum is biasing the mean neutron energy high relative to that for fission neutrons. The most likely impurities that would produce a high energy (alpha,n) neutron are magnesium, beryllium, or boron. The latter two have very high (alpha,n) neutron yields. Samples containing such impurities in the presence of plutonium are generally poor candidates for multiplicity counting and should be assayed via destructive analysis or calorimetric methods. Based on the detected heterogeneity in one can from this drum, calorimetry is the preferred assay technique.

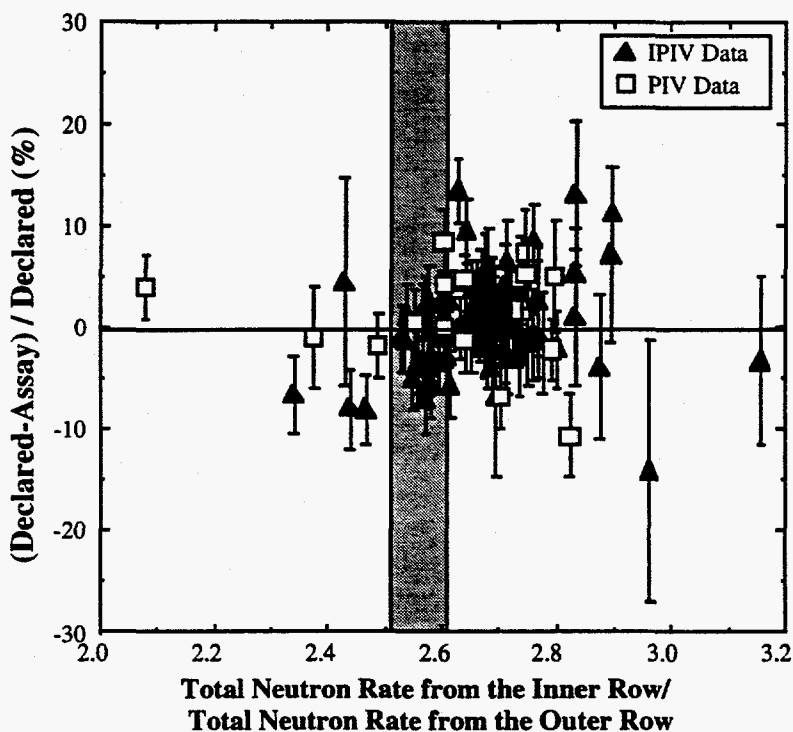


Fig. 7. Neutron rate "row ratios" showing the effect of impurities on the mean neutron energy. The shaded area is the range for pure oxide.

Conclusions

The LNMC has performed to specifications for the inventory verifications at RFETS to date. For the over 100 drums measured, the LNMC disagrees neither with operator declaration nor with destructive analysis in a statistically significant manner. Only a single drum could not be assayed with the LNMC and there is strong evidence that it contains impurities that make it unsuitable for multiplicity counting.

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