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ASSAY OF LOW-ENRICHED URANIUM USING SPONTANEOUS FISSION NEUTRONS *

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Abstract

Low-enriched uranium oxide in bulk containers can be assayed for safeguards purposes, using the neutrons from spontaneous fission of ^{238}U as a signature, to complement enrichment and mass measurement. The penetrability of the fast fission neutrons allows the inner portion of bulk samples to register. The measurement may also be useful for measuring moisture content, of significance in process control. The apparatus used can be the same as for neutron correlation counting for Pu assay. The neutron multiplication observed in ^{238}U is of intrinsic interest.

1. Introduction

The work reported here concerns an attempt to employ technology developed originally for the assay of plutonium, for two applications relating to low-enriched uranium in bulk packages.

The first of these applications is purely of safeguards interest. Low-enriched UO_2, either as feed for manufacturing or product from recovery is packaged in containers of the order of 10 kg (or more) which curently are assayed during safeguards inspection by a mass (weight) measurement coupled with an enrichment measurement using gamma-ray based techniques. The enrichment measurement assays only the surface layer of the UO_2; anything about a centimeter or two in from the surface is well shielded as regards any assay method involving gamma rays. The conventional method for assaying bulk low-enriched UO_2 thus gives no assurance that other nuclear material, e.g., high-enriched U,

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might not be clandestinely concealed within the package. The penetration of fast neutrons, either from an external source, or generated by spontaneous fission of ^{238}U, is much greater than that of gammas and in principle allows the interior of the package to be probed. The availability of neutron well counters and neutron correlation circuitry together with the seeming simplicity of using the sample's self-generated neutrons dictated trying this approach first.

The second application, measuring the moisture content of UO_2 powder, is more technologically directed, although it has some safeguards interest as well; in fact the problem was suggested by a fuel manufacturer.

Both these projects required some data relating to neutron emission from ^{238}U in order to predict the outcome of the measurements and allow comparison with expected values. The spontaneous fission half life of ^{238}U is orders of magnitude below that of the even Pu isotopes, and so not surprisingly, the corresponding data for ^{238}U is not nearly as well known.

A brief review of some of the elements of correlation counting is necessary to understand some of the features of this work and will be discussed.

The multiplication phenomenon in ^{238}U loans itself to comparatively straightforward analysis and is therefore of intrinsic interest.

2. The Spontaneous Fission Rate and the Predicted Gross Count Rate, G

Table I lists values of half-life $t_{\frac{1}{2}}$ for the spontaneous fission of ^{238}U gleaned at random from the literature. Although incomplete,[1] the list is representative of experimental results obtained in the last thirty years and indicates the degree of uncertainty with which this value has been known.

Table I

Partial List of ^{238}U Half-Life ($t_{\frac{1}{2}}$) for Spontaneous Fission

Authors	Reference	Half-Life (10^{15}y)
Perfilov	ZETP 17 476 (1947)	13 ± 2.0
Segre	Phys. Rev. 86 21 (1952)	8.0 ± 0.3
Kuroda, et al.	J. Chem. Phys. 25 603 (1956)	10.3 ± 1.0
Parker-Kuroda	J. Inorg. Nucl Chem. 5 153 (1957)	8.0 ± 0.5
Gerling, et al.	Radiokhimiya 1 223 (1959)	5.8 ± 0.5
Kuzminov	JETP 10 290 (1960)	6.5 ± 0.3
Fleischer & Price	Phys. Rev. 133 B63 (1964)	10.1 ± 0.3
Roo & Kuroda	Phys. Rev. 147 884 (1966)	8.9 ± 1.0
Roberts	Phys. Rev. 174 1482 (1968)	9.9 ± 0.2
Galliker & Hugentobler	Helv. Phys. Acta 43 593 (1970)	8.19 ± 0.06
Thury	Acta Phys. Austraica 33 375 (1971)	8 ± 0.4
Leme, et al.	Nucl. Inst. Meth. 91 577 (1971)	9.5 ± 0.2
Ivanov & Petrzhak	Sov. At. En. 36 403 (1974)	9.73 ± 0.44
Emma & LoNigro	Nucl. Inst. Meth. 128 355 (1975)	9.6 ± 0.2

Three recently derived values[1] for the spontaneous fission decay constant $\lambda_{sf}=\ell n2/t_{\frac{1}{2}}$, considered the best now available, are (8.46 ± 0.06), (8.7 ± 0.6), and (8.57 ± 0.42) in units of $10^{-17}y^{-1}$. The mean value and standard deviation of the mean from these is $\lambda_{sf}=(2.71_8 \pm 0.16_3)\times10^{-24}s^{-1}$. This value will be adopted in the following. As will be seen, the accuracy with which measured and calculated values can be compared will depend mainly on the accuracy with which λ_{sf} for ^{238}U is known.

The spontaneous fission source strength q of a mass m (grams) of ^{238}U is therefore $q = (L/A)\lambda m = (6.022\times10^{23}/238.07)(2.72\pm0.16)\times10^{-24}m = (6.88\pm0.04)\times10^{-3}m$ fissions per second, where L is Avogadro's constant and A is the nuclear mass number.

The gross count, G, of a neutron well counter due to this spontaneous fission is $G=\varepsilon\langle\nu\rangle q$, where ε is the efficiency for neutron detection of the counter averaged over the neutron energy spectrum, and $\langle\nu\rangle$ is the average value of ν,

the number of neutrons emitted per fission.

Some measured values of $\langle\nu\rangle$ for ^{238}U spontaneous fission, $\langle\nu_{sf}\rangle$, are listed in Table II.[1] An average value for $\langle\nu_{sf}\rangle$ and its uncertainty obtained from the last four entries is $\langle\nu_{sf}\rangle = 2.04 \pm 0.08$ neutrons/spontaneous fission. The neutron production from spontaneous fission alone is therefore $(6.88 \pm 0.40)(2.04 \pm 0.08) = (14.04 \pm 1.37)$ neutrons/kg s.

Table II

Values of $\langle\nu\rangle$ for Spontaneously Fissioning ^{238}U

Authors	Reference	Quoted Value
Segre	Phys. Rev. 86 21 (1952)	2.2 ± 0.3
Littler	Proc. Phys. Soc. (London) A65 203 (1952)	2.5 ± 0.2
Geiger & Rose	Can. J. Phys. 32 498 (1954)	2.26 ± 0.16
Richmond & Gardner	AERE R/R 2097 (1957)	2.14 ± 0.07
Kuzminov, et al.	JETP 10 290 (1960)	1.7
Gerling & Shukolyokov	Sov. At. Energy 8 41 (1961)	2.1 ± 0.1
Leroy	J. Phys. Rad. 21 617 (1960)	2.10 ± 0.08
Asplund-Nilsson, et al.	Nucl. Sci. Eng. 15 213 (1963)	1.97 ± 0.07
Conde & Holmberg	J. Nucl. Eng. 25 331 (1971)	2.00 ± 0.05

There will also be a yield of (α,n) neutrons. The production rate of these will be strongly influenced by the presence of light element impurities such as ^{19}F, and $^{17,18}O$ (in the case of UO_2), and by the enrichment (isotopic fractions of ^{235}U present), since it depends on the α emission rate.

Uncertainties in the (α,n) production rate make gross (ordinary) neutron counting an unreliable assay tool for plutonium (with exceptions). Though there is, of course, no comparable amount of experience with U or UO_2, presumably similar considerations apply. Neutron correlation ("coincidence") counting in principle separates out the spontaneous fission component from the (α,n) or, indeed, any other Poisson distributed (random) neutron background. The correlation

count will be proportional only to the number of fissions, and the only way the uncorrelated (Poisson) component will affect it is in increasing the statistical uncertainty in the correlation count.

Correlation counting[2] was therefore used as well as gross counting in this study.

3. Other Required Parameters

Prediction of the correlation count rate ^{238}U requires a knowledge of the quantity $\langle\nu(\nu-1)\rangle \equiv \sum\nu(\nu-1)P_\nu$, where ν is the number of neutrons emitted per fission, P_ν is the neutron multiplicity distribution, the probability that ν neutrons were emitted, and the angular brackets $\langle\rangle$ denote an average taken of the quantity within the brackets. (In principle the distribution P_ν would also be required in order to obtain $\langle\nu_{sf}\rangle = \sum\nu P_\nu$, but, in practice, it can be determined just by monitoring the number of fission events and the total number of detected neutrons, correcting the latter for the detection efficiency.)

There doesn't seem to be any published information on the P_ν for ^{238}U. However, using the facts that (i) the "Diven's" parameter formed by the ratio $D \equiv \langle\nu(\nu-1)\rangle/\langle\nu\rangle^2$ is relatively insensitive with respect to nucleon number, and that (ii) to the extent it does vary, it seems to be subject to systematic behavior at least in the vicinity of ^{238}U, allows one to make a reasonably good estimate for $\langle\nu(\nu-1)\rangle$. (The parameter D is a measure of the relative width of the P_ν distribution and would equal 1 if the P_ν were Poisson distributed.)

Relevant data for even (spontaneously fissioning) and odd number nuclides are listed in Table III. Other experimental data are available, but the two sets used were each derived using the same apparatus and analysis and so are more appropriate to study systematic behavior. As can be seen, $\langle\nu(\nu-1)\rangle$ and $\langle\nu\rangle$

(or $\langle\nu\rangle^2$) differ more among themselves than does the ratio D. Figure 1, a graph of D vs. nucleon number A, shows D = 0.80 ± 0.15 for A from 233 to 244 inclusive. From Figure 1, an estimate for D and its uncertainty for ^{238}U is D = 0.811±.020. Therefore, an estimate for $\langle\nu(\nu-1)\rangle$ for ^{238}U is $(0.811 \pm .020)(2.04 \pm 0.08)^2$, or $\langle\nu(\nu-1)\rangle = 3.375 \pm 0.277$. The ≈ 8% uncertainty in $\langle\nu(\nu-1)\rangle$ is mainly due to that in $\langle\nu\rangle$.

Table III

Data on the Parameter $D \equiv \langle\nu(\nu-1)\rangle/\langle\nu\rangle^2$

	$\langle\nu(\nu-1)\rangle$	$\langle\nu\rangle$	$\langle\nu\rangle^2$	D
^{252}Cf	12.344	3.82	14.5924	.8459
^{244}Cm	6.318	2.84	8.0656	.7833
^{242}Cm	5.558	2.65	7.0225	.7915
^{242}Pu	3.762	2.18	4.7524	.7916
^{240}Pu	4.102	2.257	5.0940	.8053
^{239}Pu				.815
^{238}Pu	4.398	2.33	5.4289	.8101
^{236}Pu	4.252	2.30	5.2900	.8038
^{235}U				.795
^{233}U				.786

Even isotopes data from Hicks, Ise, Jr. and Pyle, Phys. Rev., 101, 1016 (1956); data on odd isotopes (80 keV induced fission) from Diven, Martin, Taschek, and Terrell, Phys. Rev., 101, 1012 (1956).

4. *Prediction of* the Correlation (Net) Count

The basic equation[2] relating the correlation ("net") count of a *Böhnel* (shift register) type of pulse train analyzer to the parameters of the neutron well counter and of the analyzer is $N = \frac{1}{2}\langle\nu(\nu-1)\rangle\varepsilon^2 Fqt$ where $\langle\nu(\nu-1)\rangle$ is as defined above (the average being computed using the P_ν for the nuclide being assayed), ε is the efficiency for detecting a single neutron (averaged over the neutron energy spectrum), F is a quantity depending on the lifetime of neutrons

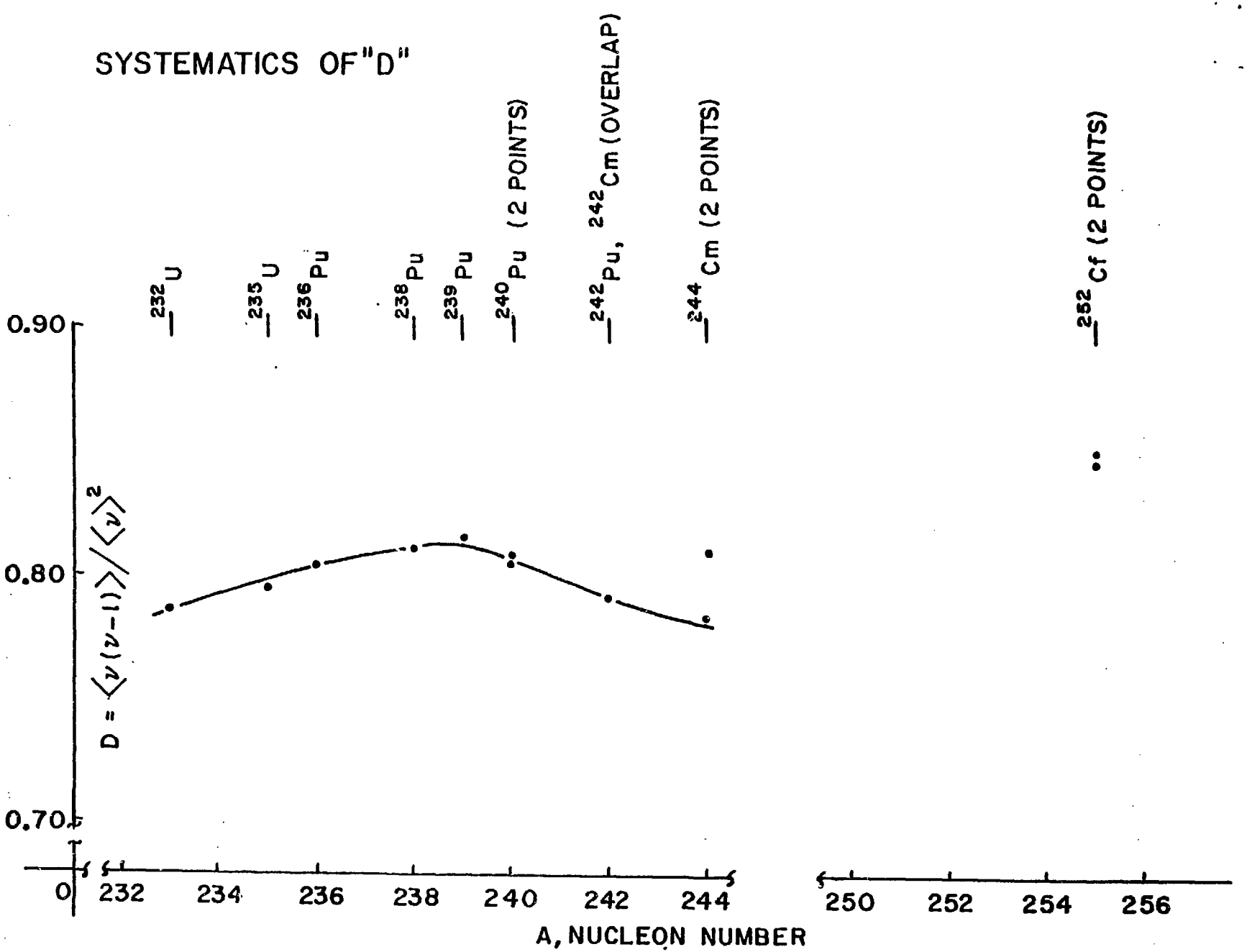
SYSTEMATICS OF "D"
^{232}U
^{235}U
^{236}Pu
^{238}Pu
^{239}Pu
^{240}Pu (2 POINTS)
^{242}Pu, ^{242}Cm (OVERLAP)
^{244}Cm (2 POINTS)
^{252}Cf (2 POINTS)
0.90
0.80
0.70
$D = \langle \nu(\nu-1)\rangle / \langle \nu \rangle^2$
0
232
234
236
238
240
242
244
250
252
254
256
A, NUCLEON NUMBER

FIG. 1

in the counter and particular parameters of the correlation circuitry, q is the number of fissions per unit time, and t is the time. The quantity F is a number between 0 and 1, typically of the order of 0.5 for a given detector and control setting of the electronics, and is given by $F = \exp(-\tau_1/\tau_o)\left(1-\exp(-\tau/\tau_o)\right)$, where τ_o is the lifetime ("die away time") of neutrons in the counter, τ_1 is a built-in predelay in the electronics ($\sim$ 4 μs), and τ is the correlation time set by the operator, generally chosen to be of the order of the detector die away time (typical values used would be between 20 and 120 μs depending on the neutron detector and other factors).

The above expression for N can be generalized for the situation with more than one nuclide being present. For a total mass m and isotopic fraction f_i (by mass) for the i th nuclide, $N = \frac{1}{2}LFtm\sum\langle\nu(\nu-1)\rangle_i\varepsilon_i^2\lambda_i/A_i$, summed over i, where L is Avogadro's number, λ_i is the decay constant for spontaneous fission, and A_i is the atomic weight in atomic mass units if m is in grams. If all the nuclides have similar energy spectra, then $(N/t) = m(\frac{1}{2}LF\varepsilon^2)S$,

$S = \sum S_i \equiv \sum \langle\nu(\nu-1)\rangle_i\lambda_i f_i/A_i$.

In the present experiment an ANL ZPPR "F" rod was used for calibrating the well counter-detector system; the parameters involved in calculating S are listed below (Table IV).

Table IV

Parameters of ZPPR "F" Rod

Even* Isotope	A_i(g)	$\lambda_i(10^{-19}s^{-1})^{-2}$	$\langle\nu(\nu-1)\rangle_i$	$f_i(10\ \ )$	$S_i(10^{-24}g^{-1}s^{-1})$
^{238}Pu	238.05	4.482	4.398	0.0437	3.619
^{240}Pu	240.06	1.877	4.102	11.536	370.0
^{242}Pu	242.06	[illegible]39	3.762	0.188	9.173

$S = 3.828 \times 10^{-22}g^{-1}s^{-1}$
(m = 11.847 g·Pu)

* The ^{238}U contribution is negligible by comparison with the even Pu isotopes.

The net count for the F rod standard was $(25.37 \pm .35)s^{-1}$. Thus for this detector plus electronics system the quantity

$(\frac{1}{2}LF\epsilon^2) = (25.37 \pm .35)/(11.847)(3.828 \times 10^{-22}) = (5.59 \pm 0.08) \times 10^{21}$.

Therefore, the net count per gram second for this system is (N/mt) = $(5.59 \pm 0.08) \times 10^{21}$ S.

For natural U, f = 0.9929, $\langle\nu(\nu-1)\rangle = (3.375 \pm 0.277)$; $\lambda = (2.72 \pm 0.16) \times 10^{-24}s^{-1}$; for ^{238}U, A = 238.07, so that for ^{238}U in this apparatus S = $(3.83 \pm 0.54) \times 10^{-26}g^{-1}s^{-1}$, and $(\frac{1}{2}LF\epsilon^2)S = (2.14 \pm 0.33) \times 10^{-4}g^{-1}s^{-1}$ = N/mt. Expressing the mass m in kg, the expected net or correlated count per second is then $(N/t) = (0.214 \pm 0.033)m$. This calibration method thus relates the performance of the system with one type of material to that with another.

The pertinent parameters of the well counter electronics system for which this calibration result pertains happens to be τ_o = 85 μs, τ_1 = 4 μs, τ = 64 μs, ε = 0.18, which are fairly typical. The sample cavity is Cd lined to prevent thermalized neutrons from returning to the sample, and possibly inducing fission in the 0.71% ^{235}U present. The parameter τ_o can be determined by plotting the

natural logarithm of the change in N versus τ for a constant source. The negative inverse of the slope is the die away time (Figure 2). This method follows from the theoretical expression for N cited above.

In order for such a calibration to be transferrable to a completely different mix of nuclides, there must be some assurance that the respective neutron energy spectra are comparable. An empirical result for the average neutron energy (in MeV) from fission is $\langle E_n \rangle = 0.74 + 0.653\ (\langle \nu \rangle + 1)^{\frac{1}{2}}$.[3] Evaluating this for ^{240}Pu (the ZPPR rod) and ^{238}U yields respectively 1.92 and 1.87 MeV. The relative difference (2.6%) would not noticeably affect the efficiency ε of a counter, the spectra having similar shapes as well.

5. Multiplication

Multiplication in ^{238}U and $^{238}UO_2$ was studied by using metal and oxide samples of natural uranium as flux multipliers for a ^{238}Pu source. This is essentially the "add-a-source" or "add-a-gram" technique.[4] The source was small (9mm D x 38 mm L), triply encapsulated, and contained ~ 0.4 g $^{238}Pu^{16}O_2$. As indicated, the oxygen content was free of $^{17,18}O$ in keeping with its intended use as a "pacemaker" source. Thus (α,n) production was minimal. The neutron production rate of this ^{238}Pu source overwhelmed that due to the spontaneous fission or (α,n) production in the ^{238}U or $^{238}UO_2$ samples themselves, though they ranged up to masses of about 9 kg. The ^{238}Pu source was counted bare in a neutron well counter, and the gross G and net N counts compared with those resulting when the source was embedded in jars of oxide, or sandwiched between slabs or chunks of metal. The ratio of G_M (the gross count rate of the ^{238}Pu source when surrounded with ^{238}U) to G (the gross count of the ^{238}Pu source alone) was defined to be the multiplication, $M \geq 1$, i.e., $G_M = M\varepsilon\langle\nu\rangle q$. Since the

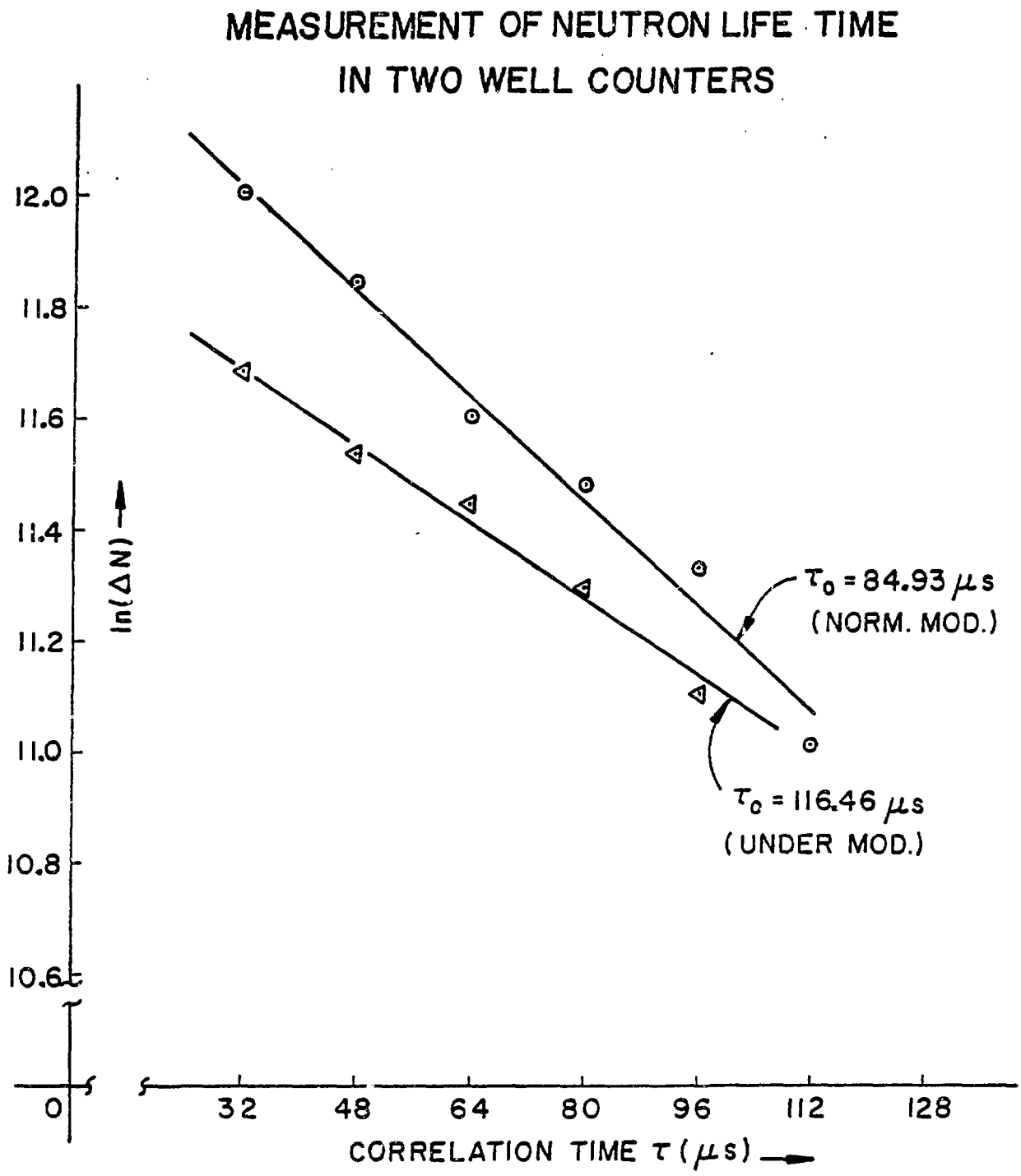
MEASUREMENT OF NEUTRON LIFE TIME
IN TWO WELL COUNTERS
12.0
11.8
11.6
11.4
11.2
11.0
10.8
10.6
ln(ΔN)
τ_0 = 84.93 μs
(NORM. MOD.)
τ_0 = 116.46 μs
(UNDER MOD.)
0
32
48
64
80
96
112
128
CORRELATION TIME τ (μs)

FIG. 2

thermalization time of fast neutrons entering the detector is of the order of, < 5μs, much less than the counter die away time (≃ 85 μs), any additional neutrons produced by fast neutrons from the ^{238}Pu source causing fission in the ^{238}U are treated by the counter-electronics system as coming from a ^{238}Pu fission with a greater multiplicity of neutron emission, namely Mν instead of ν.[5] Thus in the expression for N, $\langle M\nu(M\nu-1)\rangle \equiv M^2\langle\nu^2\rangle - M\langle\nu\rangle$ replaces $\langle\nu(\nu-1)\rangle \equiv \langle\nu^2\rangle - \langle\nu\rangle$.

Defining (N_M/N) as the ratio of the net count of the ^{238}Pu source with ^{238}U present, to the net count with the bare source, the ratio (N_M/N) may be written as: $(N_M/N) = (1+a)M^2 - aM$, where $a \equiv \langle\nu\rangle/\langle\nu(\nu-1)\rangle = 1/\langle\nu\rangle D$.

For ^{238}Pu, $\langle\nu_{sf}\rangle = \sum\nu P_\nu = 2.330$ while $\langle\nu(\nu-1)\rangle = \Sigma\ \nu(\nu-1)P_\nu = 4.398$. (The same set of P_ν was used for each calculation for consistency; probably the best quoted value of $\langle\nu\rangle$ is 2.28 ± 0.08.) Thus $a = 0.529_8$ and $(N_M/N) = 1.530\ M^2 - 0.530M$.

Values for M were obtained from the gross count data and used in the above to obtain $(N_M/N)_{CALC}$ listed in Table V and compared with the experimental values. The agreement is seen to be quite good, giving confidence in both the workings of the correlation circuitry and the model used to explain the effect of multiplication on the correlation count, N.

Table V

Effect on the N Count Due to Multiplication of ^{238}Pu Source by ^{238}U

	Chem. Form	^{238}U(kg)	G*	N*	$M \equiv (G_M/G)$	$(N_M/N)_{EXPT}$	$(N_M/N)_{CALC}$	Δ_{REL}
0**	-	0	244,762	17,528	1.000			
1	UO_2	.694	256,526	19,711	1.048	1.125	1.122	+.003
2	UO_2	.708	254,224	19,743	1.039	1.126	1.099	+.024
3	UO_2	3.78	271,867	22,094	1.111	1.260	1.297	-.029
4	U met.	2.18	276,530	23,495	1.130	1.340	1.352	-.012
5	U met.	4.45	286,288	25,791	1.170	1.471	1.471	+.000
6	U met.	8.90	303,907	29,915	1.242	1.707	1.698	+.009

$$\Delta_{REL} \equiv \left[(N_M/N)_{EXPT} - (N_M/N)_{CALC}\right]/(N_M/N)_{EXPT}$$

* Counts in 1000 s.
** ^{238}Pu source alone.

The multiplication M itself can also be modeled. The probability that a neutron from a ^{238}Pu fission goes a distance r in the ^{238}U without reacting at all is $\exp(-\Sigma r)$, with the macroscopic fission cross section given by $\Sigma = N\sigma_{ff}$, $N = L\rho/A$, where σ_{ff} is the cross section for fast neutron induced fission in ^{238}U, and ρ is the density of ^{238}U nuclei, g/cm^3. The probability of at least one interaction is therefore $1-\exp(-\Sigma r)$. (Since the interaction is absorption leading to fission, there can be only one interaction.) The quantity $1-\exp(-\Sigma r)$ represents the fraction of the incident neutron flux ϕ interacting with ^{238}U. For each interaction, one neutron is lost, but ν_{ff} neutrons are produced, where ν_{ff} is the number of neutrons produced in a fast neutron fission of ^{238}U. (This multiplicity increases monotonically with the bombarding energy of the neutrons; in general $\nu_{ff}>\nu_{sf}$.) Assume for now that the probability of further reactions by induced fission neutrons is negligible. Then, an incident flux ϕ becomes $\phi\exp(-\Sigma r) + \phi\left(1-\exp(-\Sigma r)\right)\nu_{ff} = \phi\left\{1+\left(1-\exp(-\Sigma r)\right)(\nu_{ff}-1)\right\}$ which shows the incident

flux to be multiplied by $M = 1+(1-\exp(-\Sigma r))(\nu_{ff}-1)$, in this model.

One simple refinement to this model would consider the possibility of a fraction f of neutrons from induced fission causing further fission, etc., all of these causing further additions to the flux. An infinite decreasing term geometric series is thus generated, of which the above expression for M is essentially the first term of the expansion of the usual expression for the sum of such a series.[6] However, since the fraction f is small, the new expression for M will give essentially the same result.

The average value of ν_{ff} must be calculated by considering ν_{ff} as a function of neutron energy, E_n, over the distribution of neutron energies characteristic of the fast neutron fission of ^{238}U, (using only those neutron energies above the effective threshold for fission, E_{Th} = 1.45 MeV),[7] and over the cross section for fast neutron fission, σ_{ff}. Equivalently, the product $\nu_{ff}\sigma_{ff}$ may be averaged over the relevant energy range. The value $\langle\nu_{ff}\sigma_{ff}\rangle$ = 1.56 barns has been found, appropriate to ^{238}U and $^{238}UO_2$.[8] Using $\langle\sigma_{ff}\rangle$ = 0.549 barns[8] gives $\langle\nu_{ff}\rangle$ = 2.84, to the degree of accuracy warranted.

An approximation is needed to find an "average" distance r traversed in the ^{238}U by neutrons. The volume of ^{238}U is thought of as being replaced by a sphere with the same density and volume as the original. If the density is ρ and m is the mass, then the equivalent volume of the sample is $V = m/\rho$, and the radius r of a sphere of this volume is $r = (3V/4\pi)^{1/3} = (3m/4\pi\rho)^{1/3}$, which will be taken as the mean distance traversed by neutrons from the ^{238}Pu source passing out of the sample.

Using this approach, the values calculated for M_{CALC} can be compared with those experimentally measured, M_{EXPT}, listed in Table VI. The agreement is gratifying considering the simplicity of this model. Figure 3 summarizes the

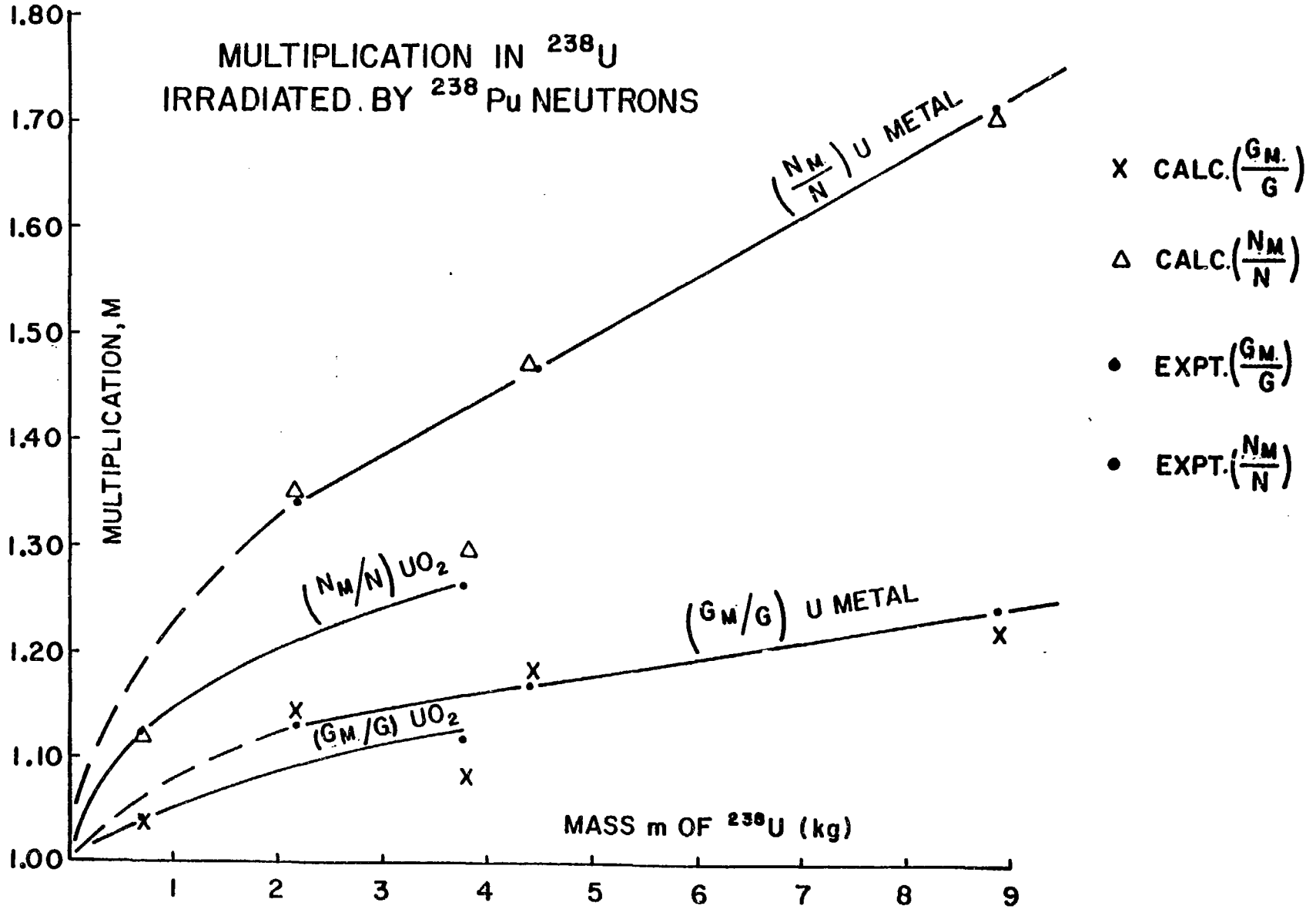
MULTIPLICATION IN ^{238}U
IRRADIATED BY ^{238}Pu NEUTRONS
MULTIPLICATION, M
MASS m OF ^{238}U (kg)
(N_M/N) U METAL
(N_M/N) UO_2
(G_M/G) U METAL
(G_M/G) UO_2
X CALC. (G_M/G)
Δ CALC. (N_M/N)
• EXPT. (G_M/G)
• EXPT. (N_M/N)
1.80
1.70
1.60
1.50
1.40
1.30
1.20
1.10
1.00
1
2
3
4
5
6
7
8
9

FIG. 3

measured and calculated values for N_M/N and G_M/G for U and UO_2.

Table VI

Effect on the G Count Due to Multiplication of ^{238}Pu Source by ^{238}U

	U mass kg	Chem. Form of U	Average Density ρ (g/cm^3)	Average Radius r (cm)	Cross Section Σ(cm^{-1})	$M=(G_M/G)_{EXPT}$	$M=G_M/G)_{CALC}$	Δ_{REL}
1	.694	UO_2	$\sim$ 4	3.61	.00491	1.05	1.03	+.019
2	.708	UO_2	$\sim$ 4	3.63	.00491	1.04	1.03	+.010
3	3.780	UO_2	$\sim$ 4	6.35	.00491	1.11	1.06	+.050
4	2.18	U met.	18.95	3.02	.0264	1.13	1.14	-.010
5	4.45	U met.	18.95	3.83	.0264	1.17	1.18	-.009
6	8.90	U met.	18.95	4.82	.0264	1.24	1.22	-.020

$$\Delta_{REL} = (M_{EXPT} - M_{CALC})/M_{EXPT}$$

A detailed computer code calculation, either transport or Monte Carlo, carried out for the actual geometry of each sample might not do significantly better. As will be seen later there may be no need for it.

6. Application to the Assay of ^{238}U

The above models may now be applied to the assay of ^{238}U with some confidence. In this case, the spontaneous fission of the ^{238}U (also, sufficiently energetic (α,n) reactions in the ^{238}U) takes the place of the ^{238}Pu source. Instead of a nearly point source, however, the neutrons now come from all over the volume of the ^{238}U sample. It might therefore seem more appropriate now to calculate M using a code. However, the approximation that the neutrons all originate at the center of an "equivalent" sphere will be used again for simplicity, to be justified by the results.

The effect on the N count is taken into account as before using the approximate constants for ^{238}U: $\langle\nu_{sf}\rangle = 2.04$ and $\langle\nu(\nu-1)\rangle = 3.37_5$, giving $a = 0.604_4$,

and $(N_M/N) = 1.604M^2 - 0.604M$.

The experimental data listed in Table VII consists of the gross count rate G_M/t and the net count rate N_M/t before correcting for multiplication. The values of the multiplication M, and the ratio N_M/N (where N is the net count if there were no multiplication) are calculated according to the method tested in the previous section. These quantities are then used to derive G/t and N/t, i.e., the gross and net count rate corrected for multiplication: $(G/t) = G_M/t)/M$ and $(N/t) = (N_M/t)/(N_M/N)$.

Table VII

Application of Multiplication Correction M to ^{238}U and $^{238}UO_2$

	Mass (kg) UO_2	Mass (kg) ^{238}U	r (cm)	M	G_M/t s^{-1}	G/t s^{-1}	N_M/N	N_M/t s^{-1}	N/t s^{-1}
1		1.42	2.62	1.123	14.5	12.9	1.344	.471	.351
2		3.03	3.37	1.157	19.0	16.4	1.447	.962	.665
3		1.79	2.83	1.132	15.3	13.5	1.372	.565	.412
4		5.87	4.20	1.193	25.4	21.3	1.562	1.970	1.261
5		3.85	3.65	1.169	21.9	18.7	1.486	1.284	.864
6	.78	.69	3.60	1.032	11.0	10.7	1.085	.189	.174
7	1.59	1.40	4.56	1.041	12.7	12.0	1.109	.382	.345
8	4.29	3.78	6.35	1.057	18.4	17.4	1.152	.996	.861
9	5.08	4.48	6.72	1.060	22.5	21.2	1.161	1.221	1.052

Since the gross count has a component to it which has nothing to do with the fission in the ^{238}U, and is impurity, background, etc., dependent, there is no theoretical prediction to compare the gross count with. The count rate predicted by the adopted value of λ_{sf} merely sets a lower limit to G. The (α,n) contribution could range from being roughly comparable (usually) to orders of magnitude greater, e.g., when ^{19}F is present.

As regards the net count, Figure 4 shows the N data uncorrected for multiplication compared to the calculated response of the well counter electronics

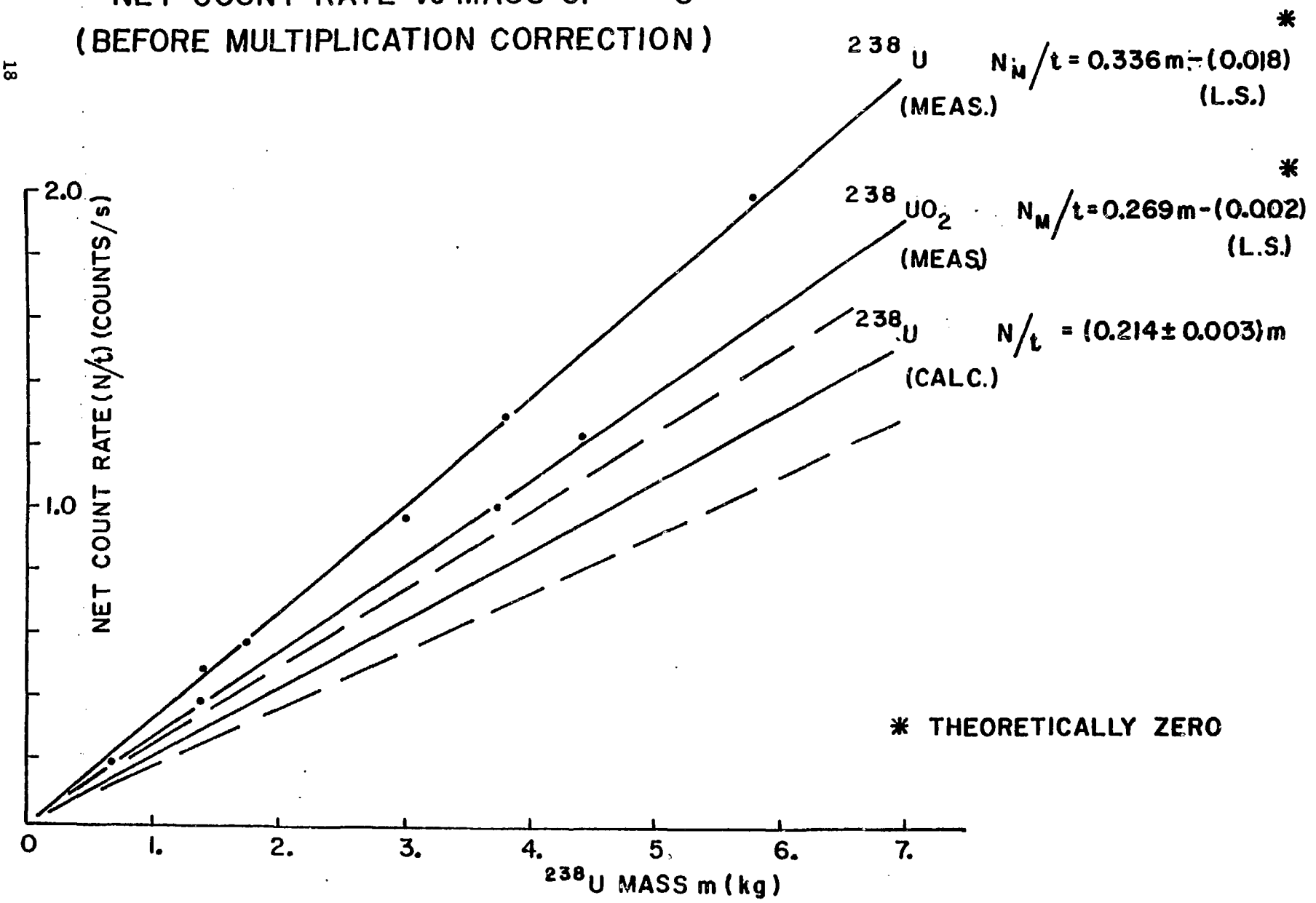
NET COUNT RATE vs MASS OF ^{238}U
(BEFORE MULTIPLICATION CORRECTION)
^{238}U (MEAS.)
$N_M/t = 0.336m - (0.018)$ (L.S.)
*
$^{238}UO_2$ (MEAS)
$N_M/t = 0.269m - (0.002)$ (L.S.)
*
^{238}U (CALC.)
$N/t = (0.214 \pm 0.003)m$
* THEORETICALLY ZERO
NET COUNT RATE (N/t) (COUNTS/s)
2.0
1.0
0
1.
2.
3.
4.
5.
6.
7.
^{238}U MASS m (kg)

FIG. 4

system. The statistical uncertainties in N range from 0.4% to 1.2%, too small to be indicated. Figure 5 shows the data corrected.

A quantitative estimate of the accuracy of the multiplication correction is furnished by a comparison of the least square (l.s.) linear fits to the uncorrected and corrected U and UO_2 data, with that for the calibration calculated on the basis of theory and the response to the ZPPR rod (Table VIII).

Table VIII

Effectiveness of the Correction for Multiplication
(Comparison of the Slopes of the l.s. Linear Fit)

Material	Meas. Slope* Before Mult. Corr.	Theoretical (Calc.) Slope*	Meas. Slope* After Mult. Corr.	Δ_{REL}
U	0.336	0.214	0.213_4	-.001
UO_2	0.269	0.214	0.230_0	+.075

$$\Delta_{REL} = \frac{\text{(corrected slope)} - \text{(theoretical slope)}}{\text{(theoretical slope)}}$$

* The units of the slope are (net) counts per kg s.

The 7.5% difference for the UO_2 may partially reflect (ironically, in view of one of the subjects of this work) the presence of moisture in these samples, which were all fairly old (probably > 20y) and stored in (otherwise unsealed) screw cap glass jars. The deviation is in the right direction, i.e., showing higher detector efficiency, as would be expected for a slightly undermoderated detector as this one was intended to be. Another factor is the crudeness with which the density had to be estimated, though admittedly, the multiplication correction is not sensitive to changes in ρ.

It is interesting to note that while the effect of the multiplication quite noticeably increases the count rate (Figure 4), the plot of N/t vs. m is still

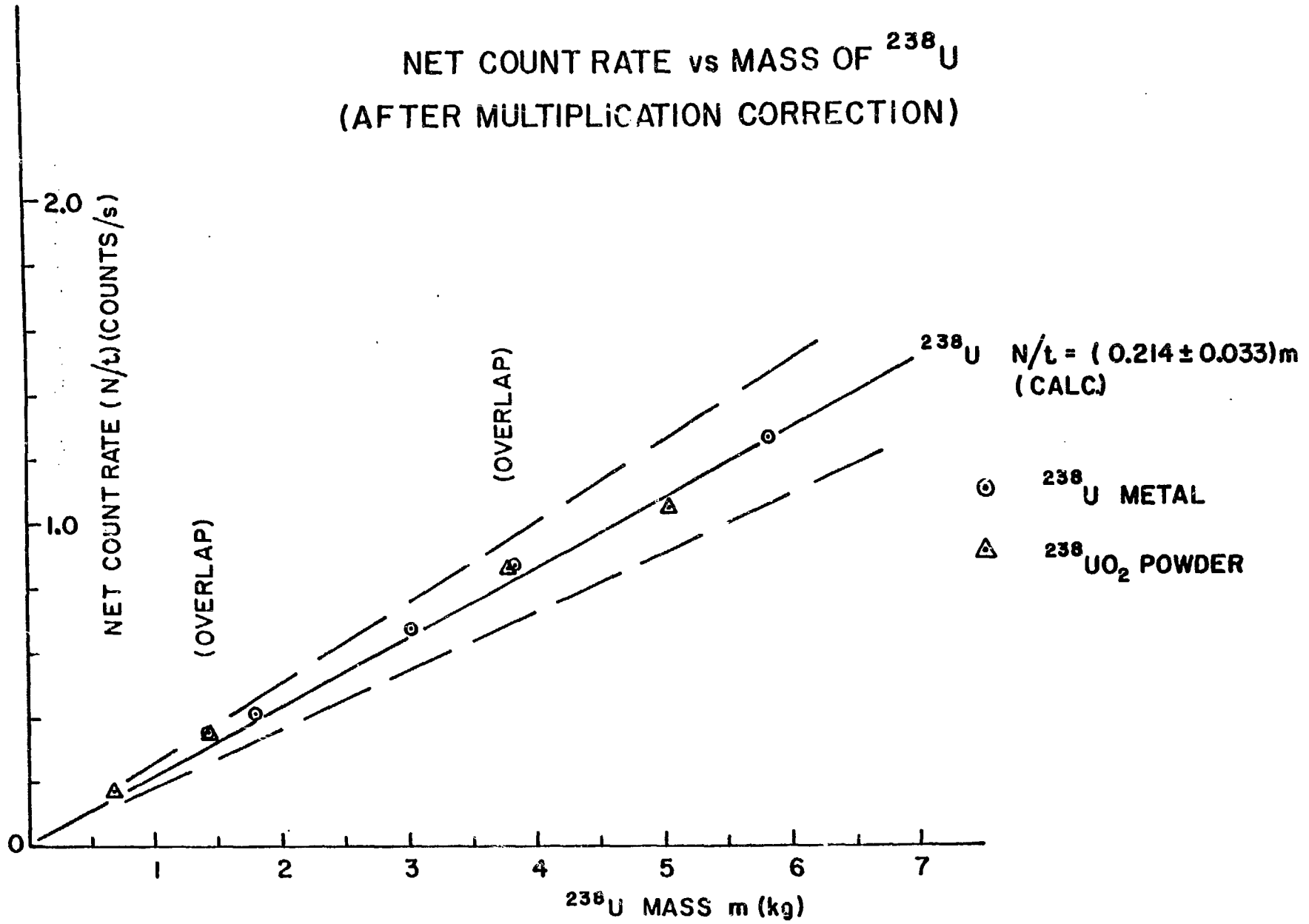
NET COUNT RATE vs MASS OF ^{238}U
(AFTER MULTIPLICATION CORRECTION)
NET COUNT RATE (N/t) (COUNTS/s)
2.0
1.0
0
(OVERLAP)
(OVERLAP)
^{238}U N/t = (0.214 ± 0.033)m
(CALC.)
^{238}U METAL
$^{238}UO_2$ POWDER
1
2
3
4
5
6
7
^{238}U MASS m (kg)

FIG. 5

fit quite accurately with a straight line for these samples, the largest being ~ 6 kg of metal.

The agreement, at least for this range of masses, is good enough so that there is little impetus to improve the theory, which seems to have an accuracy consistent with the best data usually attained in field or even laboratory measurements. There may be a point however in trying to improve the theory just to see if present agreement is accidental. Similarly, experiments with other equipment, a larger range of masses, etc., might prove worthwhile.

There does seem to be a need for more precise values of the basic physical constants. That the present group of constants used leads to results in agreement with the mean theoretical calibration curve (Figure 5) may be fortuitous, or may indicate that the uncertainties in these constants may have been evaluated too conservatively.

That this simple multiplication theory works so well is surprising and requires explanation. The agreement is certainly helped by the fact that the interaction of neutrons to induce fission is small, i.e., $1/\Sigma$ is a fair bit larger than typical sample dimensions. For similar reasons, the correction, while theoretically a function of mass, changes so slowly in this region as to be almost a constant. The calculation of an equivalent r probably works because of this combination of facts:

(i) Gauss' theorem, applicable to the inverse square law distribution of uncollided neutron flux from a point source, implies the total probability for interaction with a uniform medium will be the same no matter where within the medium the source is located. (The surface integral of flux over a Gaussian surface conciding with the surface of the sample would be the same.)

(ii) The cross section for loss of neutrons by absorption leading to interactions other than fission is small. Therefore either a neutron arising in the ^{238}U induces fission, or it escapes the sample and becomes eligible for detection by the well counter.

(iii) The typical nuclear sample, either oxide or metal is in the form of a squat uniform density cylinder whose altitude is of the same order as a typical dimension perpendicular to the altitude. Such a geometry is roughly approximated by a sphere in many applications requiring summation or integration over the volume.

7. Measurement of Moisture Content in Powdered UO_2

The idea of using neutrons to assay for moisture content is not new. It is a standard technique for monitoring soil moisture in bore holes. The basis for it is the moderation by the moisture of neutrons from a source. This moderation enhances the detection efficiency of He^3 or BF_3 proportional counters, exposed to the neutron flux giving a count rate dependent on the moisture content.

The attempt was made to apply this idea to the assay of moisture in 10 kg sealed containers of low-enriched UO_2 powder as an alternative to opening up the containers, taking samples and performing laboratory analysis, which is labor intensive. By contrast, assaying the UO_2 in a neutron well counter has the promise of being a passive operation not requiring skilled personnel.

There are several ways of using a well counter for this, all based on the principle of affecting the detector efficiency, ε. There is, first of all, a choice to be made between using the spontaneous neutron emission from the ^{238}U as a source of neutrons, or some external source. The spontaneous neutron emission is very weak, but does away with the expense, safety, and handling problems

of an external source. The other choice to be made is whether to use gross neutron counting or correlation counting as an indication of the moisture content. The gross count has better statistical precision, but is subject to influence from background, noise pickup, and to different (α,n) production rates that might exist in different samples. Moreover, the gross count rate G varies as ε, whereas the net count N varies as ε^2; in principle, the latter would be more sensitive to moisture. The net count rate, though low, is unaffected by (α,n) production and ordinary background neutrons.

In the present work it was decided to use the spontaneous neutron production to start with. The neutron correlation instrumentation allows both G and N to be monitored simultaneously. Though an external source was not tried directly for moisture measurement, the previous section describing multiplication shows this to be promising; this will be commented on again below.

Neutron well counters intended for general applications are deliberately designed to be insensitive to the presence of moderator in the material being assayed. Basically, this is accomplished by arranging the thickness of moderator in the wall of the sample cavity to be such that small changes in this thickness (or corresponding increases in the amount of moderator in the sample) would have little effect on the gross or net count rates, there being a maximum in count rate at some thickness (dependent on the fission neutron energy spectrum). More precisely, the wall thickness is made somewhat less than that required for a maximum response, i.e., slightly undermoderated. Then increasing amounts of moderator in a series of samples will just shift the response into the region of the maximum, where the change in count will be minimized. Constructed in this manner, the normal well counter is therefore intentionally ill-suited for detecting differences in moisture content.

Two well counters were used in this study. Both were originally nominally identical in all physical dimensions and designed to have a "flat" response as described above. One was modified for the test so as to be severely undermoderated by having as much polyethylene cut away from its cavity wall as was possible without structurally weakening it. Another difference between well counters was that the "normally" moderated one had a Cd liner surrounding the cavity, while the severely undermoderated one did not. This had the effect of making the efficiencies more nearly equal (about 1.069:1.0 in favor the normal counter), though the die away times, $\sim$ 85 μs for the normal detector with Cd liner, 116 μs for the undermoderated detector without, were different. (The die-away time is not important however in this low count rate application.) Normal UO_2 in nominal 10 kg quantities with three different moisture contents, 0.06%, 1.5%, and 3.0%, was assayed in both detectors. The same set of neutron correlation electronics was used for each detector.

In Figure 6 the gross count per kg s is plotted versus the moisture content W (%) for the undermoderated (U.M.) detector. (Corresponding data for the normally moderated (N.M.) detector was discovered after the end of the experiment to be inconsistent, due to some undiscovered cause and so has not been used.) In Figure 7, the net count is similar plotted for both detectors.

The plots for G/mt and N/mt are linear over this range (considered industrially useful) within statistics. A figure of merit can be defined as the relative increase in response with increasing moisture content, e.g., $(G_o/mt)^{-1}[d(G/mt)/dW]$ or $(N_o/mt)^{-1}[d(N/mt)/dW]$, where G_o and N_o are the respective values of G and N at W=0. Evaluating these for the l.s. fitted lines, G_o/mt and N_o/mt are the respective ordinate intercepts, while d(G/mt)/dW and d(N/mt)/dW are the respective slopes. The time required for a given statistical

GROSS COUNT RATE PER UNIT MASS
G/mt
vs. MOISTURE CONTENT, W
$^{238}UO_2$

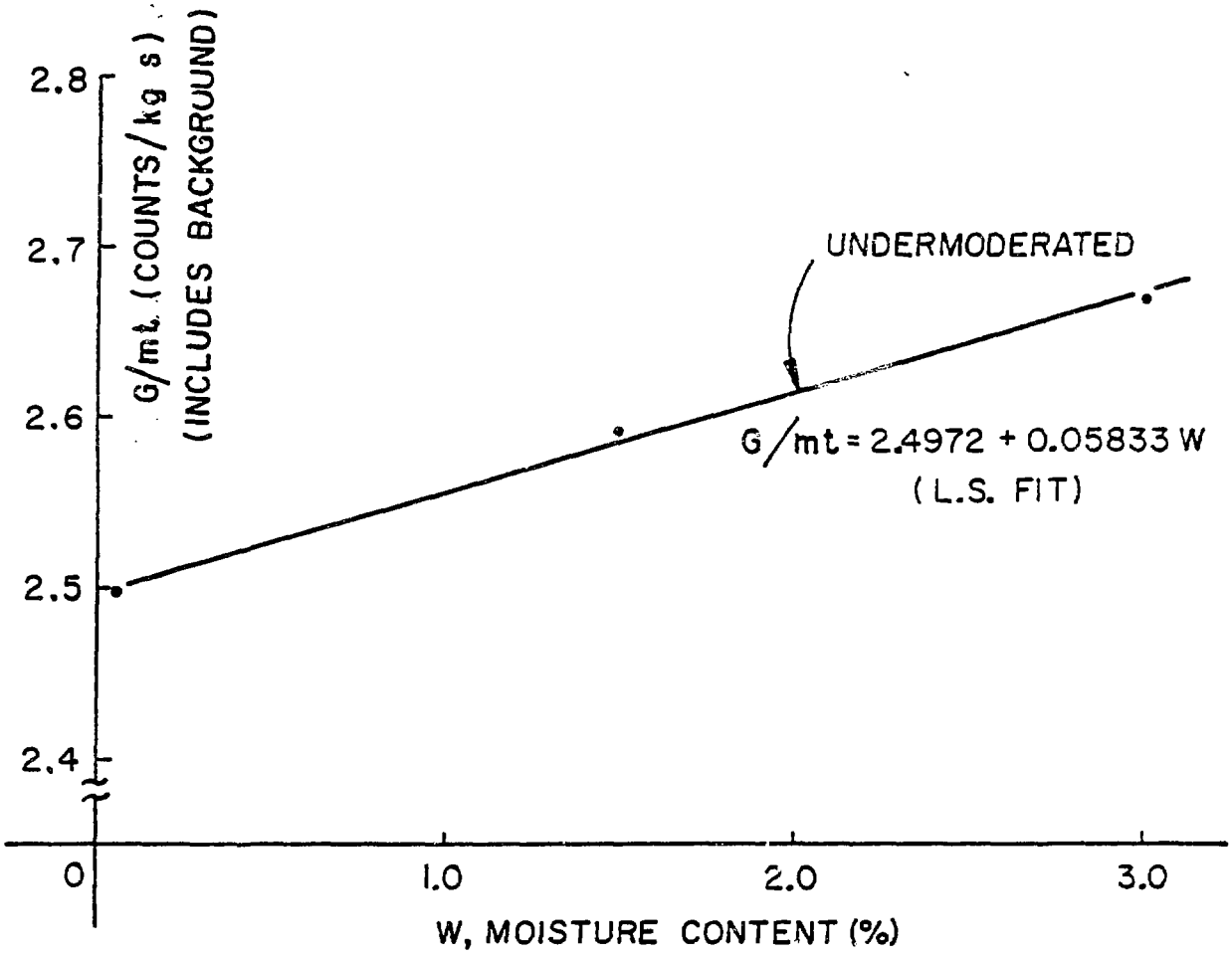

FIG. 6

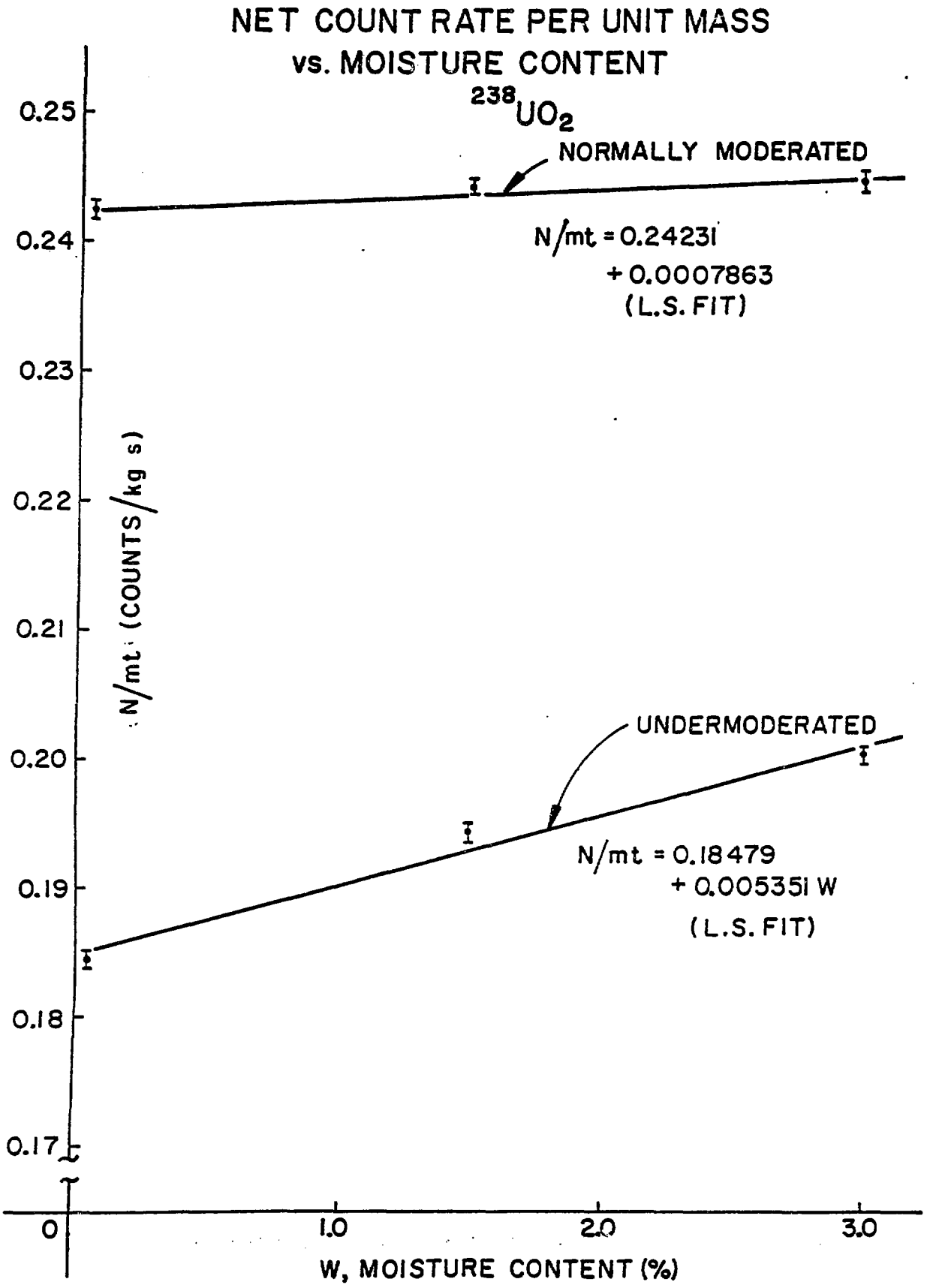
NET COUNT RATE PER UNIT MASS
vs. MOISTURE CONTENT
$^{238}UO_2$
NORMALLY MODERATED
N/mt = 0.24231
+ 0.0007863
(L.S. FIT)
UNDERMODERATED
N/mt = 0.18479
+ 0.005351 W
(L.S. FIT)
N/mt (COUNTS/kg s)
0.25
0.24
0.23
0.22
0.21
0.20
0.19
0.18
0.17
0
1.0
2.0
3.0
W, MOISTURE CONTENT (%)

FIG. 7

accuracy will vary inversely as the square root of (G/mt) $\curvearrowleft$ (G_o/mt), etc., so that reasonable overall figures of merit F_G, F_N, would be

$F_G = (G_o/mt)^{-\frac{1}{2}}\left(d(G/mt)/dW\right)$, $F_N = (N_o/mt)^{-\frac{1}{2}}\left(d(N_o/mt)/dW\right)$.

For the U.M. detector, $F_G = 0.037$, $F_N = 0.012$; for the N.M. detector $F_N = 0.0016$. From this it can be concluded that if there is assurance that the UO_2 samples are identical except perhaps for moisture content, and there is no interference from background neutrons, it would be best to use gross counting, by a factor of $\curvearrowleft$ 3:1. Otherwise, correlation counting is necessary, and in this case the use of a severely undermoderated detector is better by a factor of $\curvearrowleft$ 7.5:1.

It should be admitted that no effort was made to study the design and performance of a U.M. detector optimized for this purpose. These figures of merit may not be realistic particularly if pushed to the extremes, e.g., completely bare proportional counter tubes, which would be most sensitive to changes in moderation of the fast fission spectrum, might be too inefficient to obtain a useable count rate.

One reason this phase of the work was not pursued further was the realization that the "add-a-source" technique[4] would be far superior in count rate and could overcome some other difficulties. As mentioned previously in this paper, the sample acts simply as a way of magnifying the flux due to the introduced source. In the case of UO_2 samples, even a small source (such as a pacemaker source) will swamp out (α,n) production under reasonable circumstances, as well as the spontaneous fission components, and the sample will effectively be flooded by the source neutrons multiplied due to the sample's average macroscopic cross section for fast fission. The pacemaker source used in the present experiment was equivalent to about 100 kg of $^{238}UO_2$ as regards neutron produc-

tion. Thus, gross counting will suffice. The presence of moisture will modify the spectrum of incident source neutrons so that they will be less effective in inducing fission. It is doubtful, though, that this competing process would just cancel the increase in count rate due to moderation. Since, in an industrial situation, all the containers will be nominally identical, there will be no difference in multiplication except what is due to moisture.

Another way of analyzing the utility of this method is to consider the statistical precision attainable in the time allowed for assaying a single item. Then this standard deviation may be divided into the total change in response over the range of variation of moisture content, giving the change in W equivalent to one standard deviation. In the case of the U.M. detector the change in (N/mt) over the range (0-3)% moisture is $\curvearrowleft$ .016 net counts/kg s. The average value for σ is $\curvearrowleft$ 0.4% or $\curvearrowleft$ 8×10^{-4} net counts/kg s, achieved in about 8 hours of counting. Thus the range of (N/mt) is $\curvearrowleft$ 20 times bigger than the standard deviation. Allowing a 2σ (95%) confidence limit for adequate discrimination, approximately 0.3% absolute differences in moisture content could be seen after an $\curvearrowleft$ 8 hour count period.

An eight hour count period is not necessarily prohibitive when the alternative is considered. A package selected for moisture assay in the conventional way must be opened (breaking a tamper seal), in a hood, a representative sample must be drawn, brought to a laboratory, inserted into the apparatus for test, then returned to the hood, added to the original container, which is resealed after weighing, and returned to storage. A trail of paper (transfer documents) accompanies this process, which is clearly labor intensive.

By way of contrast, using a neutron counter involves merely inserting the sealed package into the well, pressing the start button, coming back at the end

of the run, and reading a calibration chart (or a pre-calibrated readout). Handling and paperwork are minimized. If the desired throughput cannot tolerate the eight hour wait, then either several well counters can be used in parallel, or the add-a-source technique employed. The latter could spped up throughput by an order of magnitude for even a small source (~ 1g ^{240}Pu or equivalent).

8. Conclusions

The small but measurable spontaneous fission rate of ^{238}U can be used as an assay tool. In fact, the stability and accuracy of modern correlation circuitry and ancillary electronics, and of the detector itself, allow a precision in these measurements better than that with which the fundamental physical parameters involved in the fission process are known. This points to a need for more experiments to evaluate these constants.

The fact that multiplication is a much simpler situation in the case of ^{238}U compared to Pu allows a simple model to be explored and the relation between the theory of operation of the neutron correlation circuitry and multiplication to be studied, with benefits to both.

The use of neutron measuring equipment for monitoring moisture in powdered oxides can probably be extended to PuO_2 and mixed oxides. The use of an "add-a-source" technique would be a more practical way of measuring moisture in UO_2 than using the spontaneous fission neutrons.

9. Acknowledgements

There are many people and organizations without whose help this project could not have been undertaken or would have had its scope severely limited. They are in roughly chronological order: N.E. Holden of the (U.S.) National Nu-

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10. References

1. A complete list with 35 entries spanning the year 1940 to 1976 inclusive has recently been prepared and is now undergoing evaluation at the National Nuclear Data Center (NNDC) at Brookhaven National Laboratory. The three values cited in the text, considered the best values, are respectively from D. Galliker, E. Hugentobler, and B. Hahn, Helv. Phys. Acta 43 (1970) pp. 593-606; G.A. Wagner, et al. Geochim, Cosmochim. Acta 39 (1975) pp. 1279-1286; and K. Thiel and W. Herr, Earth Planet. Sci. Lett. 30 (1976) pp. 50-56. The values of $\langle\nu\rangle$ were also furnished by the NNDC but have not been evaluated yet. The particular selection used is that of the authors.

2. For the theory and practice of modern neutron correlation equipment and well counters, see for example, M.S. Zucker, "Neutron Correlation Counting for NDA", Proc. American Nuclear Society, May 15-17, 1978. Other reports are R. Sher, "Operating Characteristics of Neutron Well Counters," BNL-50332, Brookhaven National Laboratory, (Long Island, New York, USA) (1972); K. Böhnel, "Die Plutoniumbestimmung in Kernbrennstoffen mit der Neutronenkoinzidenzemthode," (Karsruhe, FRG), KFK 2203, Gesellschaft für Kernforschung M.B.H. (1975); both of these have extensive references to work done at BNL, LASL, Karlsruhe, and other institutions.

3. J. Terrell, Phys. Rev. 113, 527 (1959) and Phys. Rev. 127, 880 (1962).

4. These techniques were first suggested and practiced by the LASL Safeguards Group. The principal names involved with the "add-a-gram" technique are M.M. Thorpe and D.B. Smith; with the "add-a-source" technique, H.O. Menlove and R.B. Walton. It is not clear at this writing whether their work ever appeared in published literature. The more pertinent LASL reports are LA-4029-MS, pp. 5-7 and 14-17, July-Sept. 1968; LA-4315-MS, pp. 8-10, July-Sept. 1969; and LA-4457-MS, pp. 26-34, Jan.-Apr. 1970. The add-a-source method was used in neutron well counters, as in this experiment, while the add-a-gram method was used in active interrogation systems. (D. Reilly, private communication).

5. This approach to the effect of multiplication on N closely parallels the work of Dowdy, et al. at LASL: "New Neutron Correlation Measurement Techniques for Special Nuclear Material Assay and Accountability," IAEA-SM-231/69, Vol. 2, Proc. Symp. "Nuclear Safeguards Technology," 1978. The treatment by Ensslin, mentioned in Krick, et al., pp. 60-62 of IAEA-SM-231/50 in the above volume, is somewhat different but perhaps could be used as well.

6. This method for calculating M is due to Spinrad, Fleishman, and Soodak, as reported in J.R. LaMarsh, pp. 402-406, Nuclear Reactor Theory, Addison-Wesley (1966).

7. A recommended source for $\langle\nu\rangle$ is R.J. Howerton, Nuc. Sci. and Eng. 62, pp. 438-454 (1977).

8. Fleishman and Soodak, Nuc. Sci. and Eng. 7, p. 217 (1960).