MLM-MU-68-65-0011

EVALUATION OF LOW ASSAY Pu-238 OXIDE FOR USE IN FABRICATION OF PLASMA-FIRED MICROSPHERES

May 15, 1968

RECEIVED
JUL 01 1996
OSTI

R. D. Madding, Jr.
R. E. Vallée

MONSANTO RESEARCH CORPORATION
A SUBSIDIARY OF MONSANTO COMPANY

MOUND LABORATORY
MIAMISBURG, OHIO
OPERATED FOR
UNITED STATES ATOMIC ENERGY COMMISSION

U.S. GOVERNMENT CONTRACT NO. AT-33-1-GEN-53

DISCLAIMER

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

EVALUATION OF LOW ASSAY Pu-238 OXIDE FOR USE IN FABRICATION OF PLASMA-FIRED MICROSPHERES

May 15, 1968

Compiled by: R. D. Madding, Jr.
Approved by: R. E. Vallée

Monsanto Research Corporation
Mound Laboratory
Miamisburg, Ohio

ACKNOWLEDGEMENT

The efforts of the numerous people in the Nuclear Operations Department who prepared and analyzed the microspheres are hereby acknowledged.

Introduction

The desirability of evaluating low assay plutonium-238 (~23% Pu-238) oxide was discussed with AEC personnel during a meeting at Germantown on July 6, 1967. In September, 1967 a sample of this material was furnished to Mound for evaluation. This evaluation was to be completed on a schedule which would not interfere with production of material for the SNAP programs, and was to include characterization of the oxide feed, and preparation and subsequent characterization of plasma-fired microspheres according to SNAP-27 procedures. Authorization to perform this work was received from DAO on September 1, 1967.

Fabrication of Microspheres

The Pu-238 oxide microspheres were fabricated from oxide feed received from the Savannah River Plant (SRP). The feed material was assayed by Mound Laboratory prior to microsphere preparation. About 30 grams of this feed material were used in preparing feed for the torch by the hydroxide precipitation method. This work, including high temperature sintering, was performed on a laboratory scale in the Research Building. The resultant torch feed was plasma-fired in the production torch in SM, sized by screening, and segregated to remove nonspherical particles.

These microspheres were then subjected to characterization according to SNAP-27 procedures, with the exception of the fines determination. Fines data on the small quantity of low assay microspheres obtained could not be considered comparable to that obtained on a production batch. Additional data collected on the oxide feed and the microspheres is also included in this report.

Compilation of Data

	Oxide Feed (as received from SRP)	Oxide Microspheres
Loss on Ignition (to 900°C)	1.03 %	-
Stoichiometry (O:Pu)	2.00	2.00
Melting Point	2150° ± 80°C	2345° ± 80°C
Apparent Density	-	10.33 g/cc
Crush Strength		
Average	-	140 grams
Maximum	-	311 grams
Minimum	-	37 grams
Impurities (as $\frac{\text{Metal}}{\text{Total Sample}} \times 100$)		
Ni	0.05 %	-
Fe	0.10 %	0.49 %
Cr	0.05 %	-
Al	0.07 %	0.03 %
Co	0.05 %	-
Zn	0.06 %	-
Ca	0.04 %	0.01 %
B	0.05 %	0.07 %
Mg	0.01 %	-
Si	0.02 %	0.17 %
Total	0.50 %	0.77 %

Isotopic and Actinide Analysis

Result reported as $\left(\frac{\text{Isotope}}{\text{Total Pu Metal}} \times 100\right)$

Element	Error[1]	Oxide Feed (as received from SRP)	Oxide Microspheres
Th-232	-	<0.010 wt %	<0.010 wt %
U-234	± 10 %	0.162 wt %	0.162 wt %
U-235	-	<0.010 wt %	<0.010 wt %

Element	Error[1]	Oxide Feed (as received from SRP)	Oxide Microspheres
Pu-236[2]		0.6 ppm	0.6 ppm
Np-237	± 10 %	0.213 wt %	0.255 wt %
Pu-238	± 1.0 %	23.04 wt %	23.57 wt %
Pu-239	± 0.7 %	59.71 wt %	59.52 wt %
Pu-240	± 1.0 %	10.39 wt %	10.36 wt %
Pu-241	± 3.0 %	5.62 wt %	5.61 wt %
Am-241	± 10 %	0.265 wt %	0.394 wt %
Pu-242	± 5.0 %	1.09 wt %	1.06 wt %

[1] Limits of error in percent of quoted value.
[2] The Pu-236 in Pu-238 is 2.4 ppm in each case, and the limits of error are $^{+16}_{-20}$ percent (feed) and $^{+10}_{-15}$ (microspheres) of the reported values.

Stack-up	Oxide Feed (as received from SRP)	Oxide Microspheres
Calorimeter Sample Weight (after loss on ignition)	1.0336 grams	1.5225 grams
Calorimeter Wattage	0.118 watts	0.175 watts
Grams Pu-238 (by calorimetry)	0.204 grams	0.303 grams
Grams Pu (calculated from isotopic ratio and calorimeter value)	0.885 grams	1.293 grams
Percentage Pu	85.62 %	84.93 %
Grams PuO_2	1.004 grams	1.467 grams
Percentage PuO_2	97.13 %	96.35 grams
Total Impurities (including actinides)	1.23 %	2.18 %
Total Stack-up	98.36 %	98.53 %

It will be noted from the above data that the average crush strength of 140 grams does not meet the SNAP-27 specification. Since it is known that the crush strength is related to the size of the particles, a particle size measurement was made on a sample of 1667 particles. The average particle size was found to be 90 microns. From data collected on production grade $Pu^{238}O_2$ microspheres, the crush strength of this size particle is expected to be ~120 grams. It is therefore concluded that the low assay material is equivalent to the production grade material with respect to crush strength.

Oxide Feed Gamma Spectra

The gamma ray spectra obtained for the low and high energy ranges are shown in Figures 1 and 2 respectively. These spectra were obtained with a standard NaI(Tl) detector 3" in diameter by 3" long positioned at approximately one meter from the source.

There is a greater than usual thallium-208 contamination to the Pu-238 spectrum, as would be expected, since the low assay material has a higher Pu-240 and Pu-236 content than is typically found in the normal 80% enriched material. Also, the 203 keV gamma ray is more intense and broader than normal. This is due to the added contribution from the 203 keV Pu-239 gamma ray and the 208 keV gamma ray from U-237.

Oxide Microsphere Gamma Spectra

Figures 3 and 4 show the gamma ray spectra obtained on a sample of the microspheres. Figure 3 covers the low energy range and Figure 4 the high energy range . Both spectra were obtained with a standard NaI(Tl) detector 3" in diameter by 3" high positioned at approximately one meter from the source.

In comparison with spectra obtained on the normal 80% enriched material, the contribution from Pu-238 to the gamma ray spectra has decreased while the contributions from Pu-239 have greatly increased as evidenced by the prominence of the 52 keV gamma ray. Also, the 20? keV gamma ray is more intense and broader than normal. This is due to the added contribution from the 203 keV Pu-239 gamma ray and the 208 keV gamma ray from U-237.

Oxide Feed Neutron Emission and Spectrum

On March 6, 1968, the total neutron emission rate of this sample (containing 20.3 g of Pu-238) was determined to be 3.89×10^5 n/s with an absolute uncertainty of ± 7%. This is a specific yield of 1.92×10^4 n/s-g Pu-238.

Figure 5 shows the measured neutron spectra for the same 20.3 g Pu-238 sample. This spectra is quite similar to other $^{238}PuO_2$ spectra, clearly showing neutrons from the $^{18}O(\alpha,n)$ reaction and from spontaneous fission of Pu-238. The portion of the measured spectrum which may be different from some other oxide samples is the region from 1-1.5 MeV. Some oxide spectra have shown about the same neutron intensity in this region while others have shown slightly less. At this point in time it is not known whether these differences are real or due to experimental uncertainty. It is known that there are differences of 5-10% in the specific yields of different oxide sources and perhaps these differences are showing up in the spectra in the 1-1.5 MeV region.

Oxide Microsphere Neutron Emission

Neutron emission measurements were made of a sample (containing 0.70 g of Pu-238) of microspheres, with the total emission rate of this sample found to be 1.34×10^4 n/s on February 23, 1968, with an absolute uncertainty of about ± 7%. The specific yield is then 1.90×10^4 n/s-g Pu-238.

Photo

Photo 1 shows a sample of the assay microspheres at about 100X magnification. The size and shape of these particles were investigated and found to meet SNAP-27 specifications.

Conclusions

The low assay (~23%) plutonium-238 oxide behaved no differently than the normal (~80%) plutonium-238 oxide throughout the feed preparation and microsphere fabrication steps of the production process. The only significant characteristic which is different from the normal material is the higher Pu-236 in Pu-238 content of the low assay material.

Based on this evaluation, it is concluded that the low assay material can be processed to acceptable product without changing the production processes.

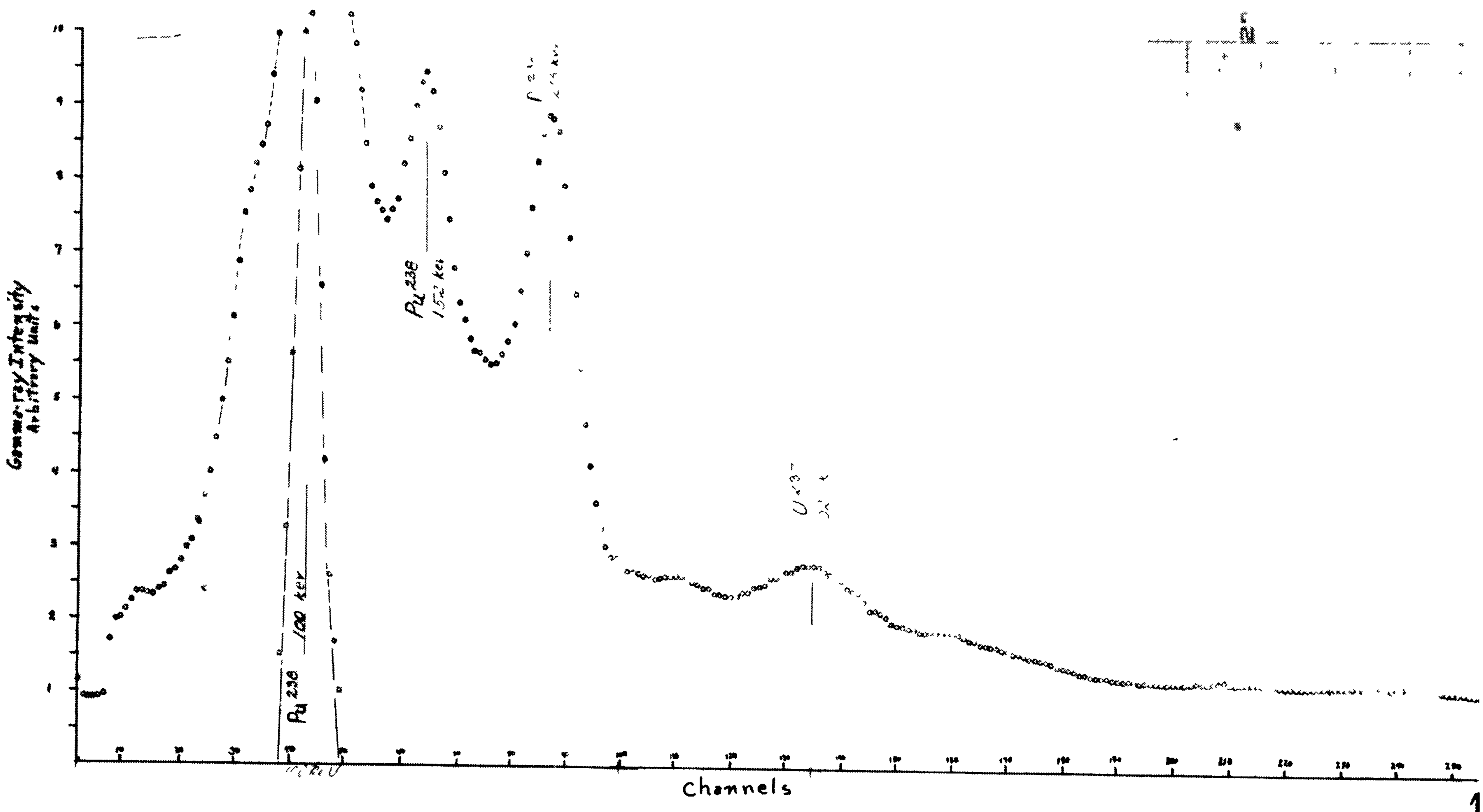

FIGURE 1

Low Assay Oxide Feed
Gamma Spectra
Range: 50 to 600 keV

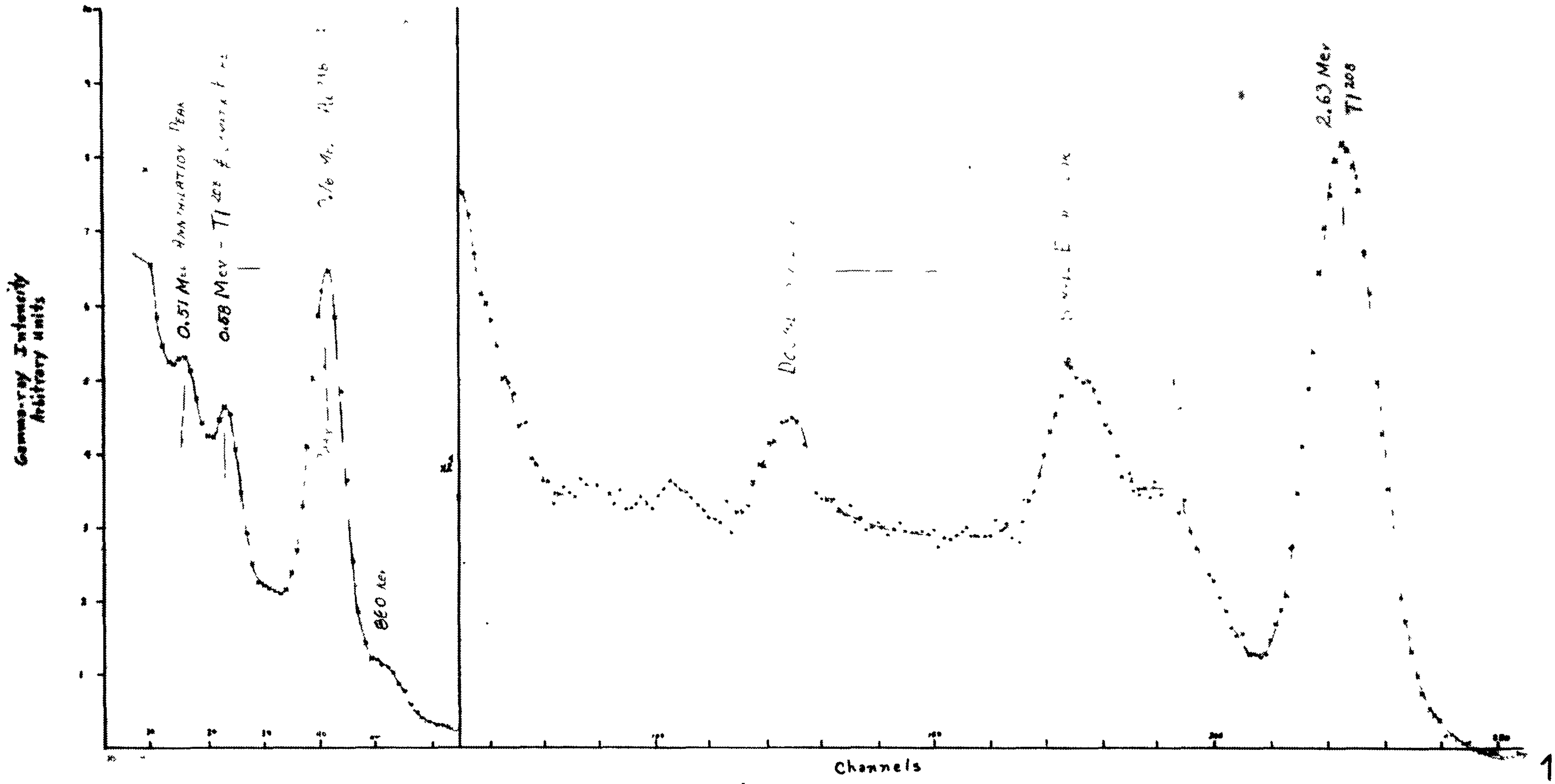

FIGURE 2

Low Assay Oxide Feed
Gamma Spectra
Range: 500keV to 600 MeV

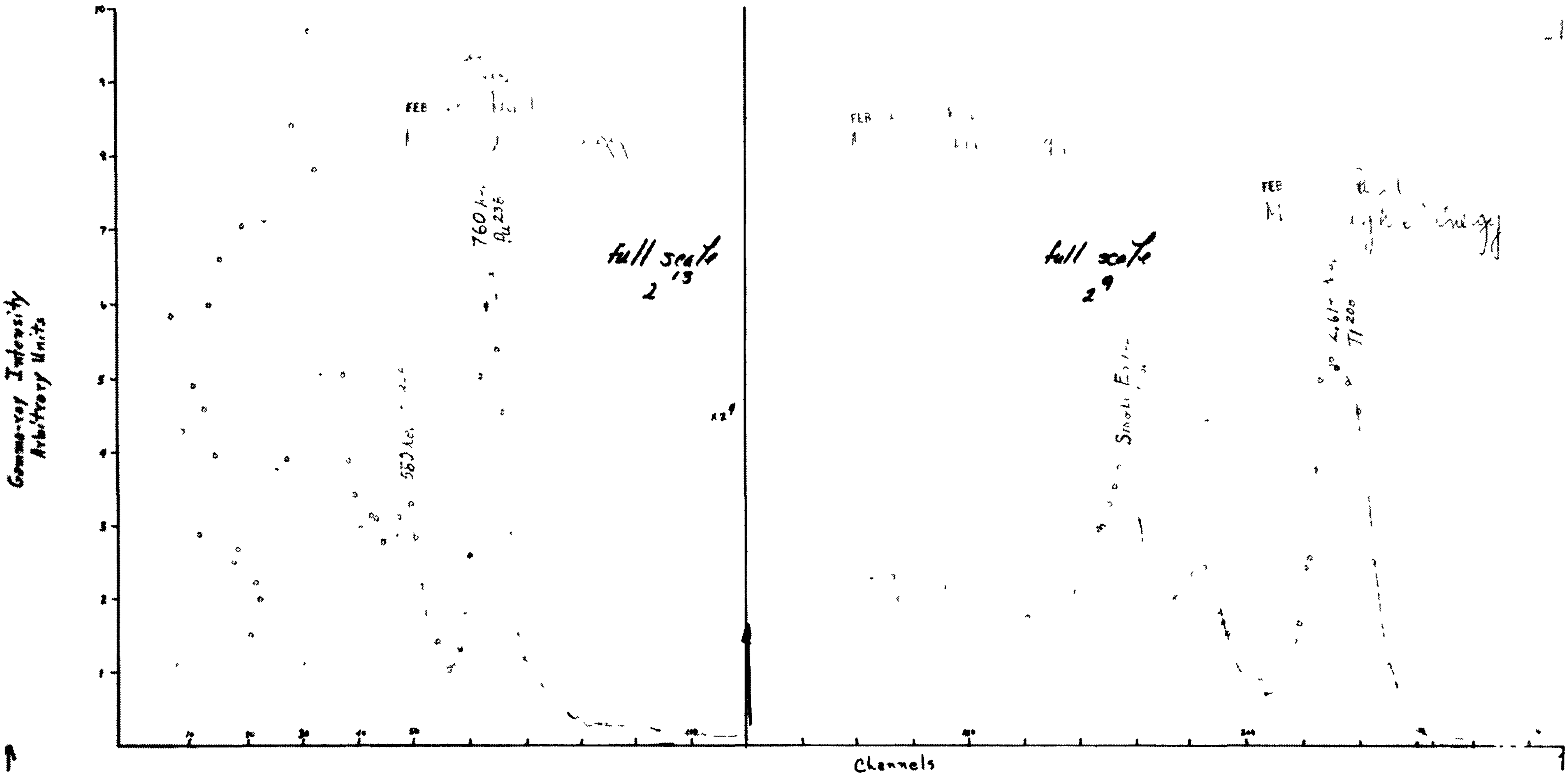

FIGURE 3

Low Assay Oxide Microspheres
Gamma Spectra
Range: 20 keV to 400 keV

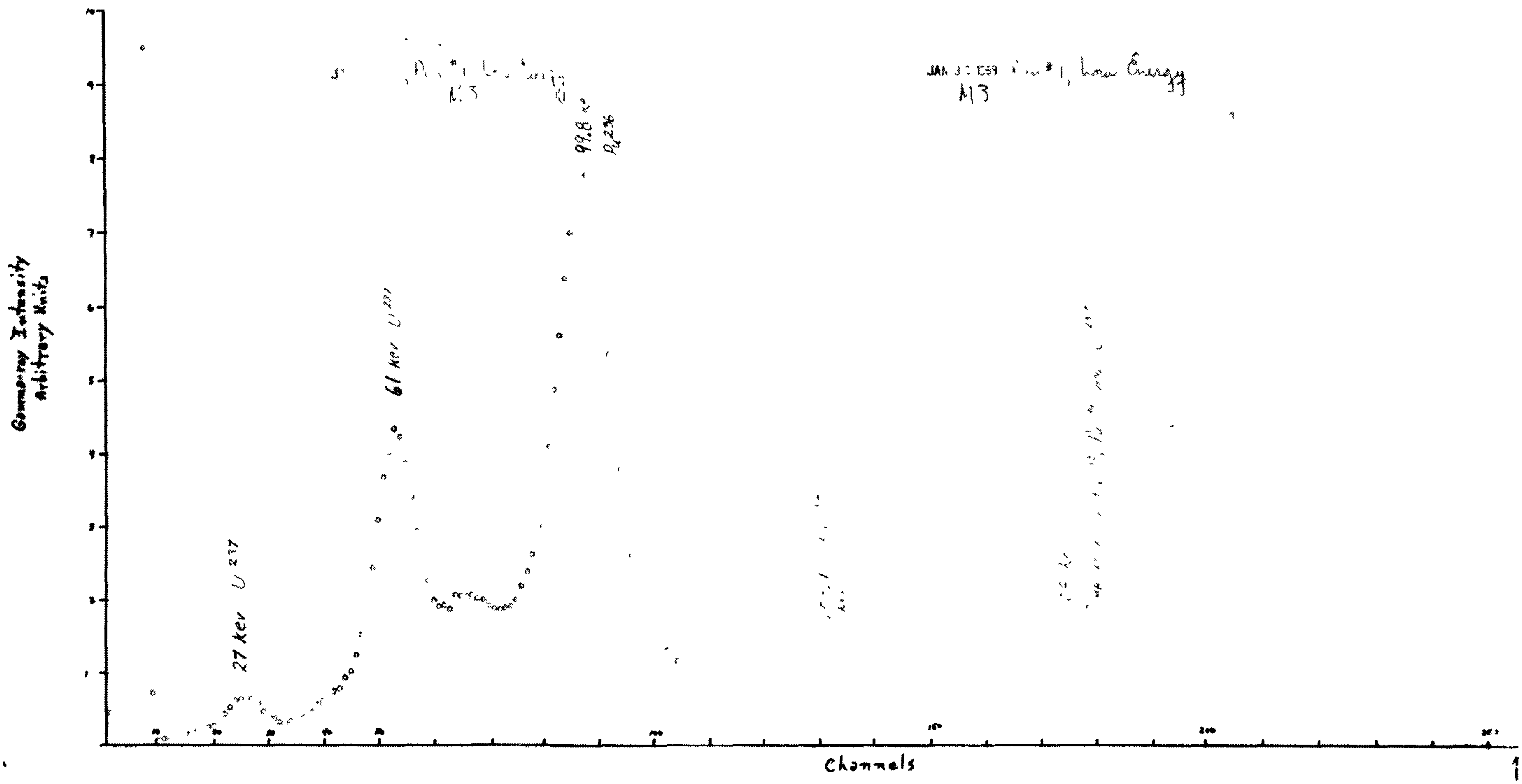

FIGURE 4

Low Assay Oxide Microspheres
Gamma Spectra
Range: 500 keV to 3.0 MeV

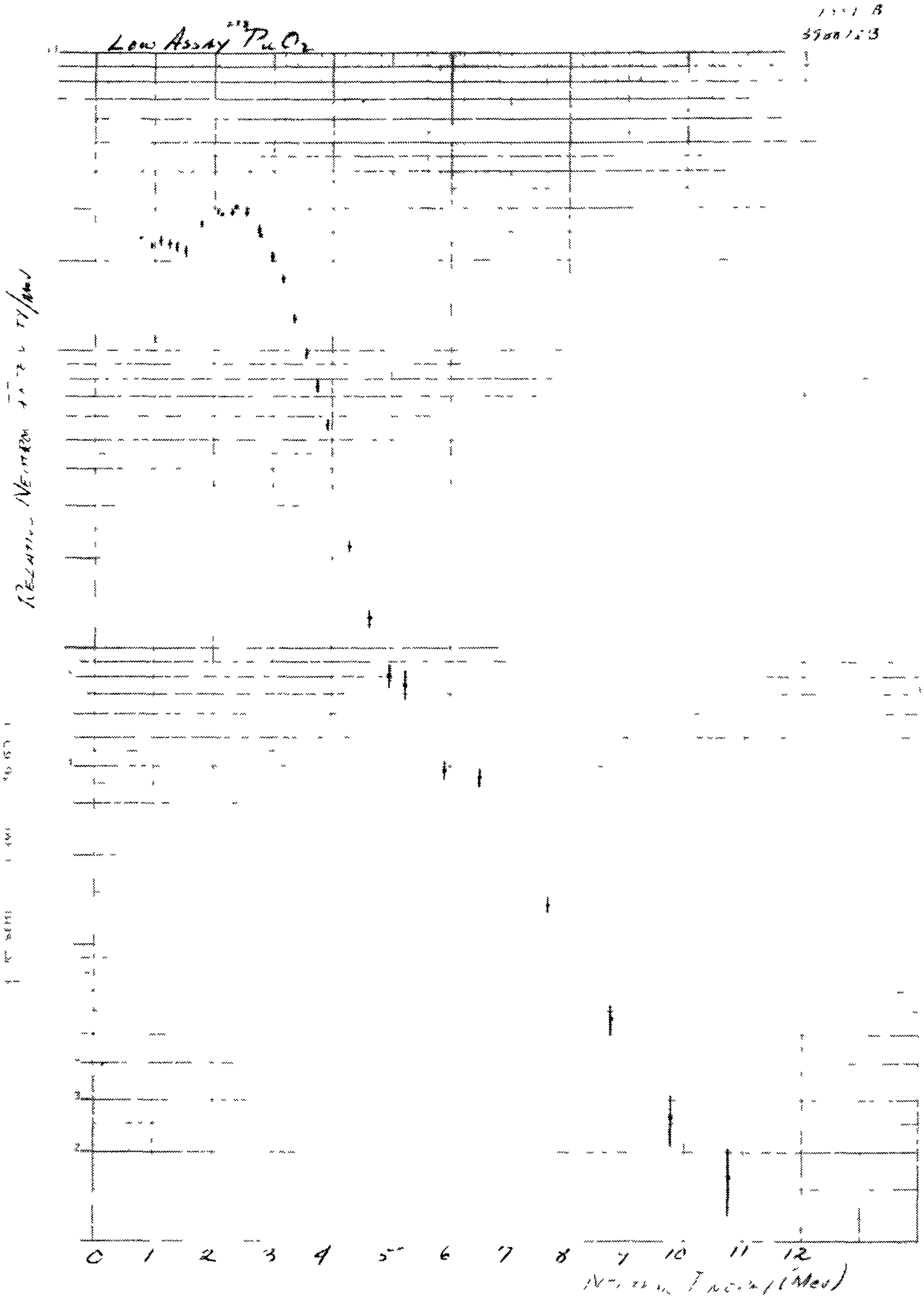

FIGURE 5

Low Assay Oxide Feed
Neutron Energy Spectra

PHOTO 1

A Sample of the Low Assay Microspheres
at about 100X Magnification