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Fertile-to-Fissile and Fission Measurements for Depleted Uranium Bombarded by 800-MeV Protons

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ABSTRACT

Axial distributions of fertile-to-fissile conversions (^{238}U to ^{239}Pu) and fissions have been measured for a thick depleted uranium target bombarded by 800-MeV protons. We determine the ^{239}Pu production by measuring the amount of ^{239}Np produced. We integrate the axial distributions to get the total conversions and fissions occurring in the target. Our preliminary experimental results give 3.81 ± 0.19 ^{239}Np atoms produced per incident proton and 5.59 ± 0.56 fissions per incident proton. Corresponding calculated results are 3.46 ± 0.05 and 3.93 ± 0.06. The computations did not include the effects of high-energy fission competition with evaporation. We also report measured axial distributions of ^{237}U and eleven fission products produced in the target. Our preliminary experimental data give 0.95 ± 0.05 ^{237}U atoms made per incident proton.

INTRODUCTION

As part of the Fertile-to-Fissile Conversion (FERFICON) program[1,2] at the Los Alamos National Laboratory, we have measured (in a thick target of depleted uranium bombarded by 800-MeV protons): a) ^{239}Pu production, b) fission, and c) ^{237}U formation. We determine ^{239}Pu production by measuring the amount of ^{239}Np formed. Other laboratories[3,4] have made similar measurements (at proton energies < 800 MeV) by observing both the radial and axial distributions of the products of interest. Our experimental approach differs significantly (as suggested by one of the authors - J. S. Gilmore) in that we combined the depleted uranium foils to integrate the product formation radially, and only explicitly measured the axial distribution for the products of interest. This substantially reduced the number of samples to be counted, and the number of gamma-ray spectra to be analyzed. We integrate the measured axial distributions over the target to obtain the total number of each reaction.

The data described here are relevant to spallation neutron source development, accelerator breeder technology, and validating computer codes used in these applications (model evaluation). We will compare our experimental data with calculated predictions using the following Monte Carlo codes: a) the Oak Ridge National Laboratory (ORNL) code HETC[5] for particle transport $\gtrsim$ 20 MeV, and b) the Los Alamos code MCNP[6] for neutron transport $\lesssim$ 20 MeV. The present Los Alamos version of HETC does not include fission when predicting particle production from nucleon and pion collisions with a fissile nucleus. A version of HETC which accounts for this high-energy fission process in uranium will soon be released by ORNL[7]; Los Alamos has requested this version of HETC from ORNL.

The 800-MeV proton source is the Clinton P. Anderson Meson Physics Facility (LAMPF).[8] We conduct the experiments at the Weapons Neutron Research facility (WNR)[9] -- see Fig. 1. We describe here our experimental setup, show some preliminary results, and compare some of our data with calculated predictions.

EXPERIMENTAL SETUP AND PROCEDURES

The location of this 'conversion' experiment in the WNR beam channel is illustrated in Fig. 2. We used a 37-rod clustered target as shown in Fig. 3. The physical characteristics of the target are given in Table I. The axial distributions of ^{239}Np, 11 fission products, and ^{237}U (a spallation product) were determined from 171 (3.239-cm diam by 0.0062-cm thick) depleted uranium foils. Nineteen weighed and matched ($\sim$1 g) foils were placed in each of seven planes perpendicular to the target axis and on the front and back target faces (see Fig. 4). Each of the 9 planes contained one foil in the central rod and 18 foils loaded symmetrically in 3 of the 6 target sectors (see Fig. 3). We chose this loading for mechanical reasons and to minimize any effects from misaligning the proton beam, which was focused on the central rod.

After an irradiation of 4.3×10^{15} protons, we prepared nine solutions for counting by dissolving the foils in hydrochloric and nitric acids. For each plane, a representative sample was obtained by mixing one-half the solution of the central-rod foil with the solution of the remaining eighteen foils. Five percent (5.00 ml) of each combined solution was used for gamma-ray counting.

To determine the number of protons striking the target, we placed a 0.0254-cm-thick Al monitor foil (sandwiched between two 0.00254-cm-thick Al guard foils to compensate for recoil losses) ~ 65 cm in front of the target. We used the number of $^{27}Al(p,x)^{7}Be$, $^{27}Al(p,x)^{22}Na$, and $^{27}Al(p,3pn)^{24}Na$ reactions occurring in the monitor foil to determine the incident proton dose. The guard foils were sufficiently thick (~ 7 mg/cm^2) to compensate for ^{7}Be, ^{22}Na, and ^{24}Na recoil losses from the central monitor-foil.[10] The number of protons determined from each of these reactions is given in Table II; we ultimately use the $^{27}Al(p,3pn)^{24}Na$ reaction in our normalizations because the cross section for this reaction is known best.[10] We located the proton beam center from the discoloration of a cellophane foil that covered one of the Al guard foils, and measured the proton beam profile by counting concentric rings cut from one of the guard foils (see Fig. 5).

All samples were counted using a Ge(Li) detector and associated pulse height analyzer, which had been calibrated against a mixed radionuclide gamma-ray reference standard.* The gamma-ray spectra were analyzed by the GAMANAL computer program.[11] After combining data from five or more counts by the CLSQ computer code,[12] we calculated the atoms of each nuclide formed using the specific gamma-rays, absolute intensities, and half-lives listed in Table III. Two of the isotopes (^{105}Rh and ^{147}Nd) had to be resolved from interfering activities by decay. We corrected all nuclide production for decay during irradiation and for gamma-ray attenuation in the sample.

EXPERIMENTAL RESULTS AND CONCLUSIONS

In Figs. 6-8, we show the measured number of atoms produced per proton per gram of uranium for eleven fission products. The shape of these (unnormalized) fission mass-yield curves do not change appreciably from front to back of the target. This latter point is further illustrated in Fig. 9 where we show the ratio of each fission product to ^{99}Mo as a function of axial position.

As shown in Fig. 10, the apparent fission-yield curve from this experiment resembles the known ~ 14.7-MeV-neutron fission of ^{238}U.[13] In Fig. 10, we compare this latter curve with our (average) measured value for each fission product. We obtained our yields by ratioing our measured

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* Amersham Corporation solution number R9/270/46.

values to that of ^{99}Mo; our 'best' estimate of the ^{99}Mo absolute yield is $(5.7 \pm 0.5)\%$. We selected our ^{99}Mo yield by envisioning curves whose summation would be noticeably greater or lower than 200% if the ^{99}Mo yield were as high as 6.2% or as low as 5.2%, respectively. The axial distributions of the fissions (based on a ^{99}Mo yield of 5.7%), ^{239}Np (^{239}Pu precursor), and ^{237}U are shown in Fig. 11 and tabulated in Table IV. The axial fission distribution is an important practical consideration (from an energy deposition viewpoint) when designing a uranium target for a spallation neutron source,[14] while the peak in the axial distribution of ^{239}Np is an aid in locating moderators to maximize low-energy ($\lesssim 1$ eV) neutron production from a uranium target.

The total number (per proton) for each reaction is

$$\text{Total}_i = \frac{M}{\ell \cdot p} \int N_i(z)dz \quad , \tag{1}$$

where M is the mass of the target in grams, ℓ is the target length in cm, p is the number of protons, and N_i is the number of atoms produced per gram of uranium. We evaluated the integral (over the target volume) using Simpson's rule. Table V lists the measured ^{239}Np, fission, and ^{237}U production per incident proton from the 30.46-cm-long target. In Table V, we also show some calculated predictions for two target lengths; the longer target was used in the Los Alamos FERFICON water-bath measurements.[1,2] In these preliminary computations, we mocked-up the rod geometry of the target exactly (exclusive of the Al target canister, etc.), and used a Gaussian proton beam spot with a standard deviation of 0.35 cm and 0.26 cm for the shorter and longer targets, respectively. When high-energy fission[15] is neglected in the computation, our preliminary comparisons between experiment and calculation show: a) that neutron production is underestimated by $\sim 10\%$, and b) that the number of fissions is underestimated by $\sim 42\%$. Note that the calculated quantities do not change appreciably for the 40.64-cm target compared to the 30.46-cm target. We are still evaluating our data and hope to report a few more fission and spallation product distributions in a final publication. For comparison with experimental data, we will calculate the axial distribution of the various products and the total product formation in the target. We will make a similar measurement using a thorium target in the near future.

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Butler for advice and assistance in counting and the use of GAMANAL and CLSQ computer programs, I. Binder for processing the Al monitor foils, and all the counting room personnel in the Los Alamos Nuclear Chemistry group. E. R. Whitaker's help in preparing the illustrations is greatly appreciated.

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TABLE I
PHYSICAL CHARACTERISTICS OF DEPLETED URANIUM TARGET

DENSITY (g/cm^3)	DIAMETER (cm)	LENGTH (cm)	^{235}U CONTENT (wt %)
19.04	19.704[a]	30.460	0.251

[a]Effective diameter ($D = d\sqrt{n}$) for a 37-rod clustered target with an individual rod diameter of 3.2393 cm.

TABLE II
Al MONITOR FOIL DATA

REACTION	CROSS SECTION USED (mb)	MEASURED NUMBER OF PROTONS	STATISTICAL COUNTING ERROR (%)
$^{27}Al(p,x)\,^{7}Be$	5.7	4.25×10^{15}	±0.9
$^{27}Al(p,x)\,^{22}Na$	13.6	4.30×10^{15}	±1.1
$^{27}Al(p,3pn)\,^{24}Na$	10.8	4.32×10^{15}	±0.3

TABLE III
NUCLEAR PARAMETERS USED TO CALCULATE
NUCLIDE YIELDS

ISOTOPE	HALF LIFE (DAYS)	GAMMA ENERGY (keV)	INTENSITY (γ/d)
^{95}Zr	64.05	756.71	0.546
^{97}Zr	0.704	743.36	0.926
^{99}Mo	2.767	140.51	0.8014
^{103}Ru	39.45	497.08	0.8637
^{105}Rh	1.473	319.4	0.1960
^{112}Pd	0.838	617.4	0.4289
^{115}Cd	2.208	336.23	0.459
^{132}Te	3.25	954.56	0.1659
^{140}Ba	12.8	487.03	0.4463
		1596.18	0.9540
^{143}Ce	1.375	293.26	0.4130
^{147}Nd	11.04	531.10	0.1310
^{237}U	6.75	208.0	0.2170
^{239}Np	2.35	277.6	0.1387
^{7}Be	53.29	477.6	0.103
^{22}Na	949.6	1274.6	0.9995
^{24}Na	0.625	1389.2	1.000

TABLE IV
MEASURED ^{239}Np AND ^{237}U PRODUCTION AND NUMBER OF FISSIONS IN THE DEPLETED URANIUM TARGET

FOIL POSITION	DISTANCE FROM TARGET FRONT FACE (cm)	^{239}Np PRODUCTION (ATOMS/PROTON·GRAM)[a]	^{237}U PRODUCTION (ATOMS/PROTON·GRAM)[a]	NUMBER OF FISSIONS (FISSIONS/PROTON·GRAM)[b]
1	0.00	1.69×10^{-5}	6.75×10^{-6}	3.65×10^{-5}
2	2.51	2.78×10^{-5}	1.07×10^{-5}	5.76×10^{-5}
3	5.01	3.40×10^{-5}	1.08×10^{-5}	6.04×10^{-5}
4	7.52	3.49×10^{-5}	9.62×10^{-6}	5.55×10^{-5}
5	10.02	3.19×10^{-5}	7.82×10^{-6}	4.63×10^{-5}
6	13.03	2.84×10^{-5}	6.08×10^{-6}	3.75×10^{-5}
7	17.04	2.16×10^{-5}	4.00×10^{-6}	2.59×10^{-5}
8	22.54	1.26×10^{-5}	2.04×10^{-6}	1.38×10^{-5}
9	30.46	4.23×10^{-6}	4.79×10^{-7}	3.24×10^{-6}

[a]Nominal estimated error is ±5%.

[b]Nominal estimated error is ±10%.

TABLE V
PRELIMINARY EXPERIMENTAL DATA
COMPARED WITH CALCULATED RESULTS

	EXPERIMENT	CALCULATION[a]	
	TARGET LENGTH (30.46 cm)	TARGET LENGTH (30.46 cm)	TARGET LENGTH (40.64 cm)
^{239}Np PRODUCTION (atoms/protons)	3.81±0.19	3.46±0.05	3.71±0.05
NUMBER OF FISSIONS (fissions/proton)	5.59±0.56	3.93±0.06	4.09±0.06
^{237}U PRODUCTION (atoms/proton)	0.95±0.05	----	----

[a]The effects of fission competing with evaporation were not included in the calculations.

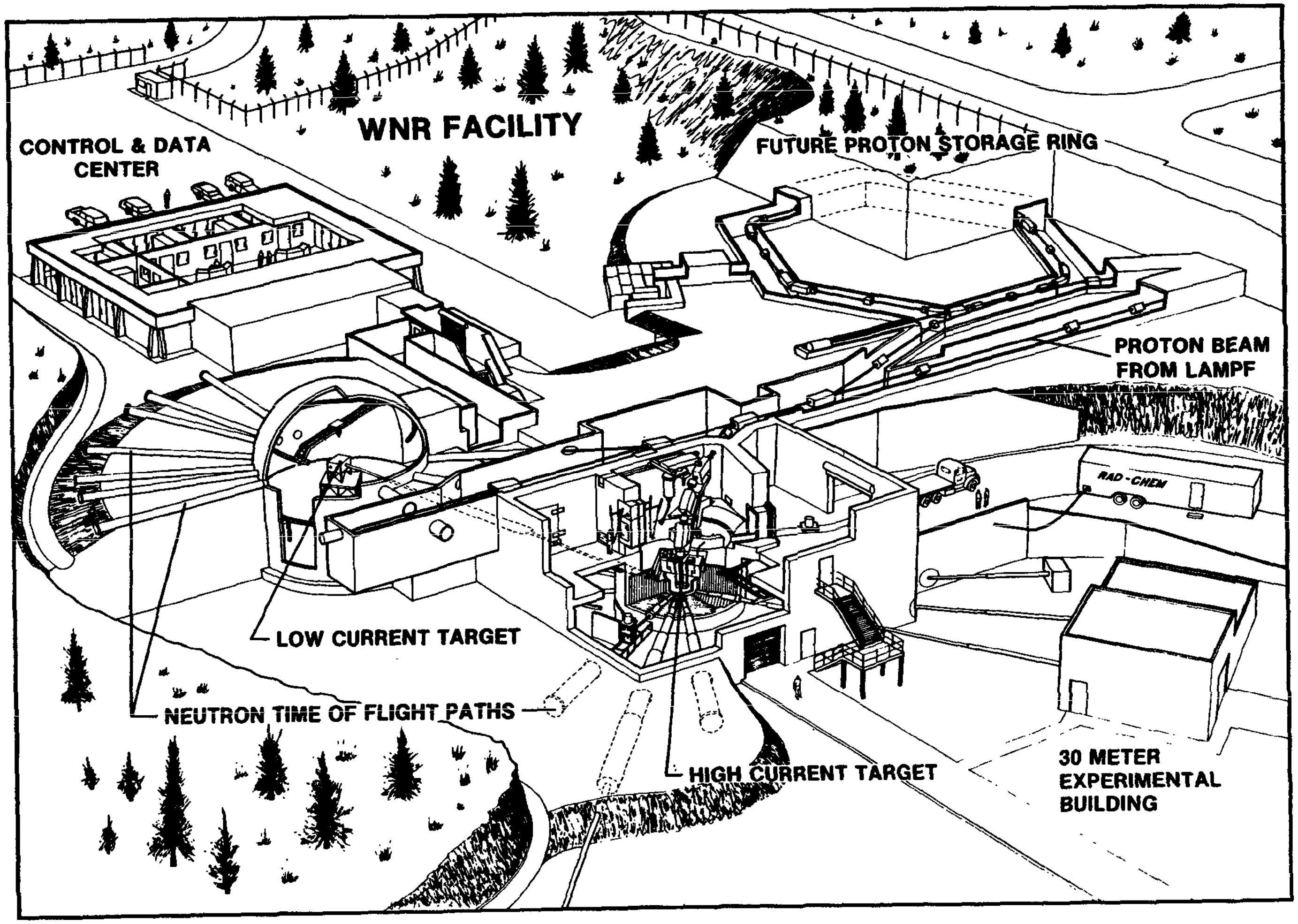

Fig. 1. General layout of the WNR showing the two target areas. The high-current target is located in a vertical proton beam and is viewed by 11 horizontal flight paths. The low-current target is located in a horizontal proton beam and viewed by 11 horizontal flight paths and one vertical flight path.

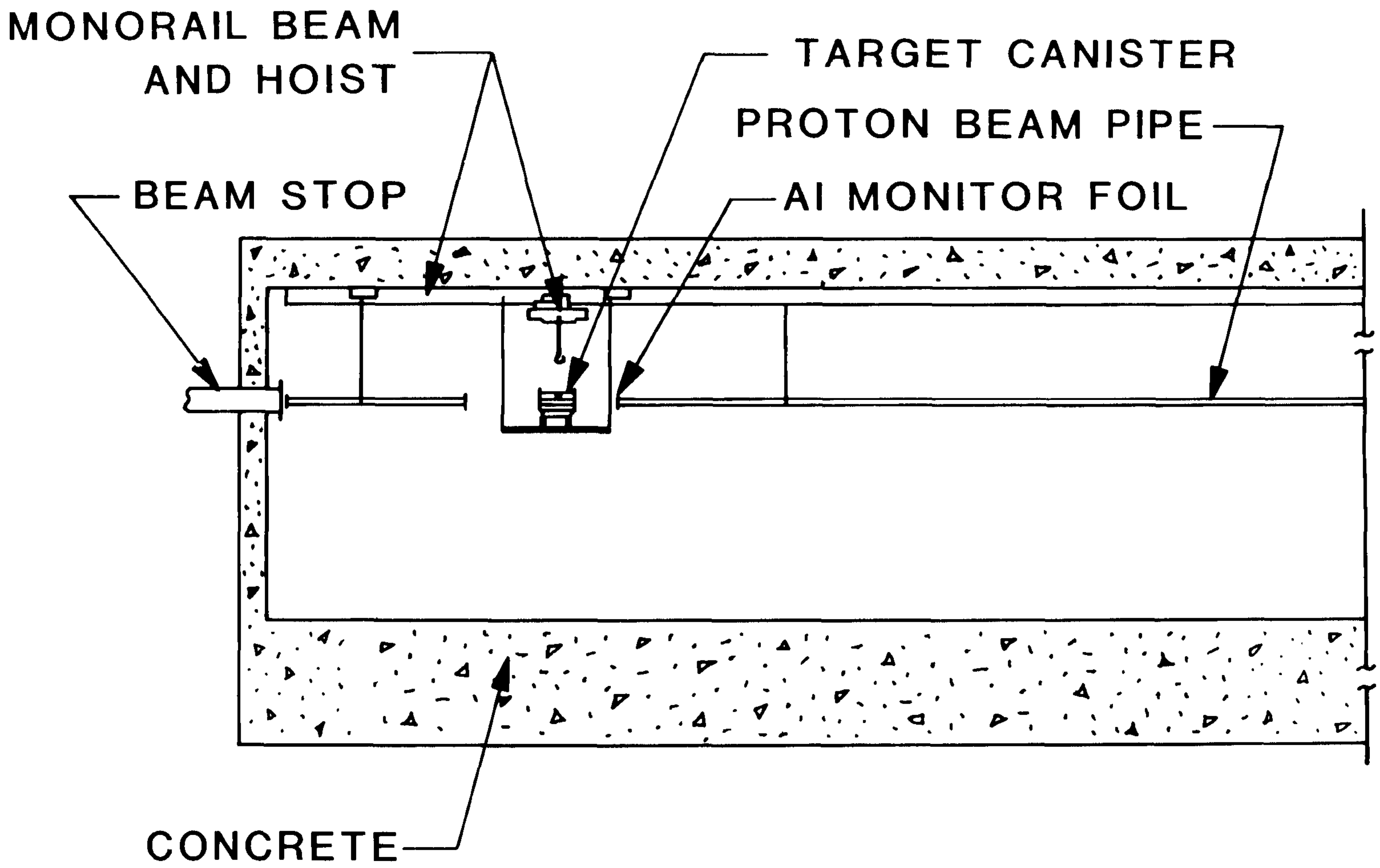

Fig. 2. Section thru the WNR beam channel showing the location of the FERFICON conversion experiment.

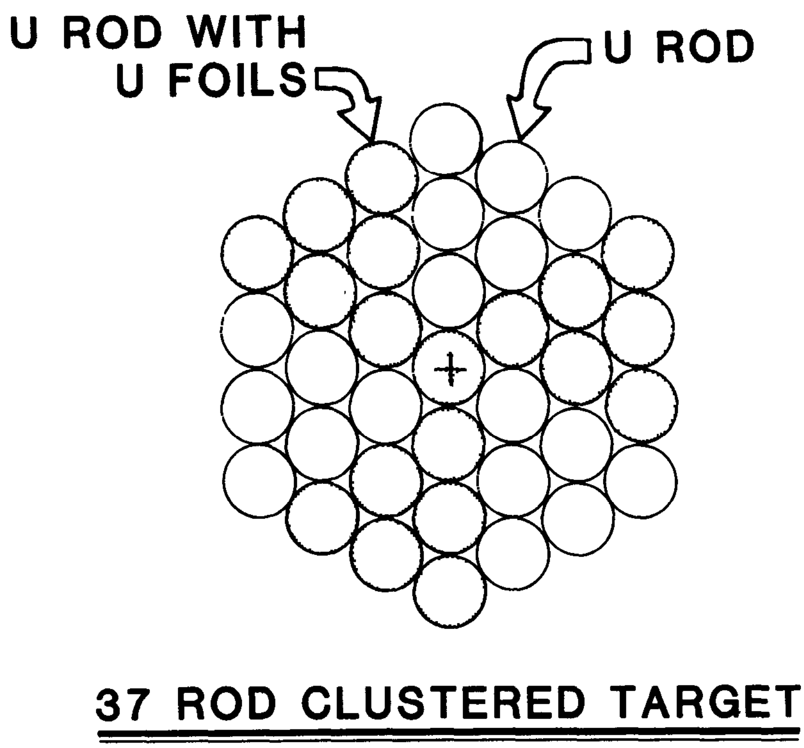

Fig. 3. Illustration of the 37-rod clustered target and the location of the foils in the array.

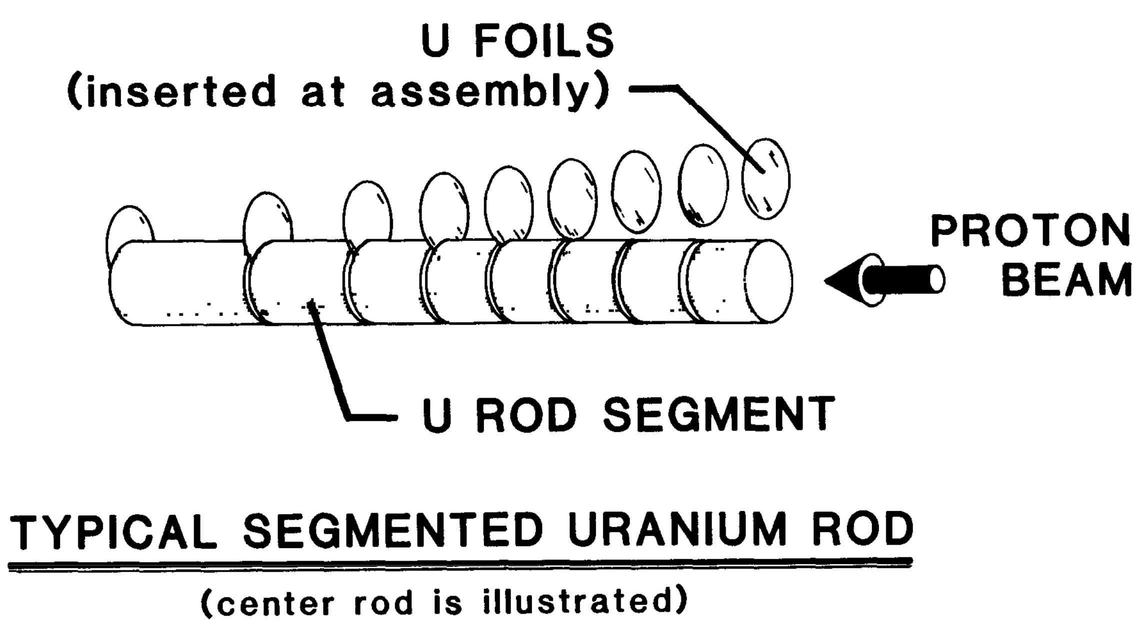

Fig. 4. Illustration showing the foil positions within a rod.

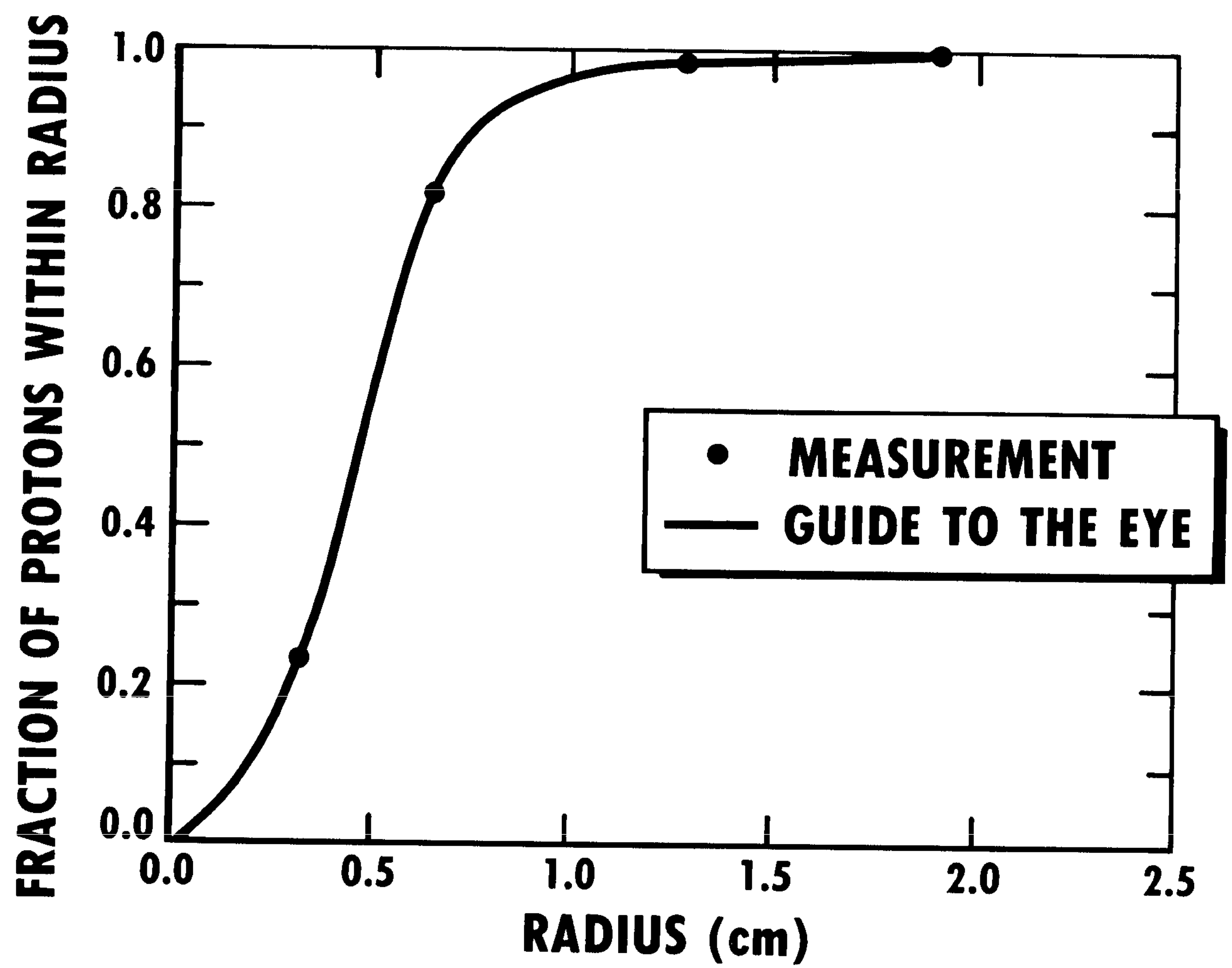

Fig. 5. Measured spatial distribution of the proton beam. The radius of the individual uranium rods was 1.62 cm.

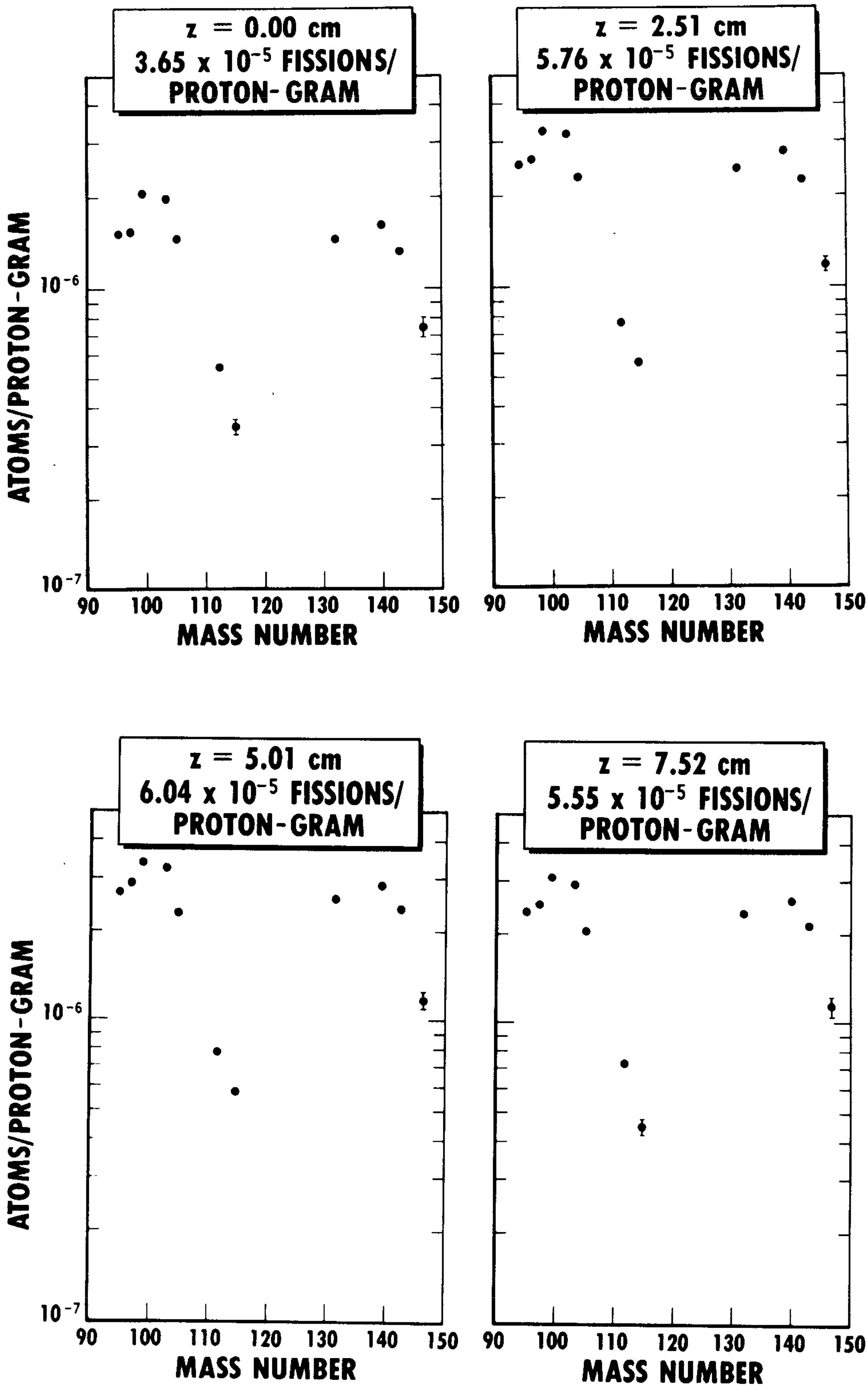

Fig. 6. Measured production of various atoms as a function of target axial position. The position z = 0.00 cm is the front target face where the 800-MeV protons were incident.

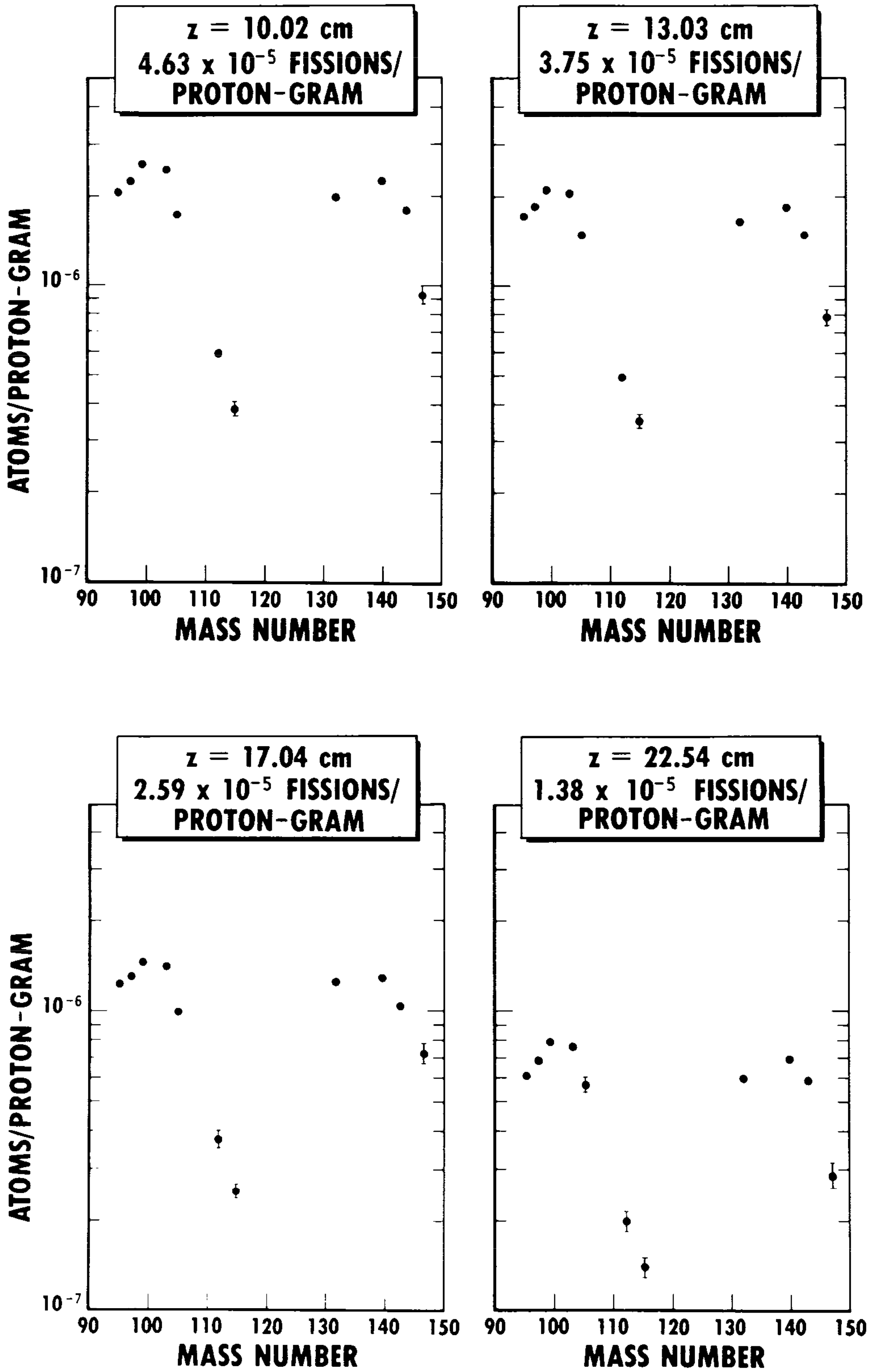

Fig. 7. Measured production of various atoms as a function of target axial position. The position z = 0.00 cm is the front target face where the 800-MeV protons were incident.

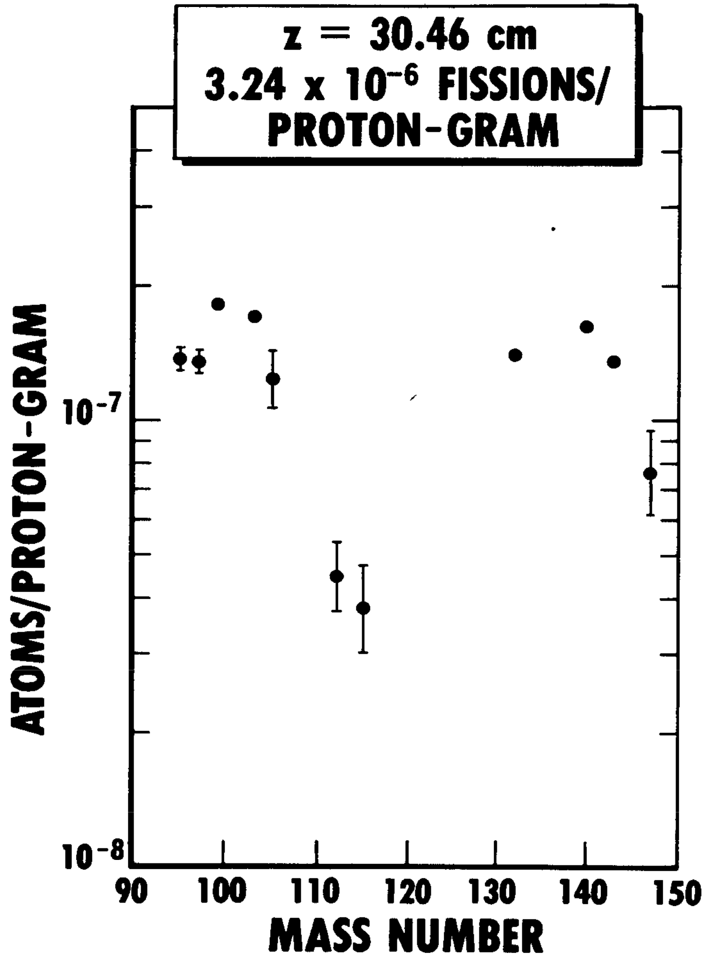

Fig. 8. Measured production of various atoms as a function of target axial position. The position z = 0.00 cm is the front target face where the 800-MeV protons were incident.

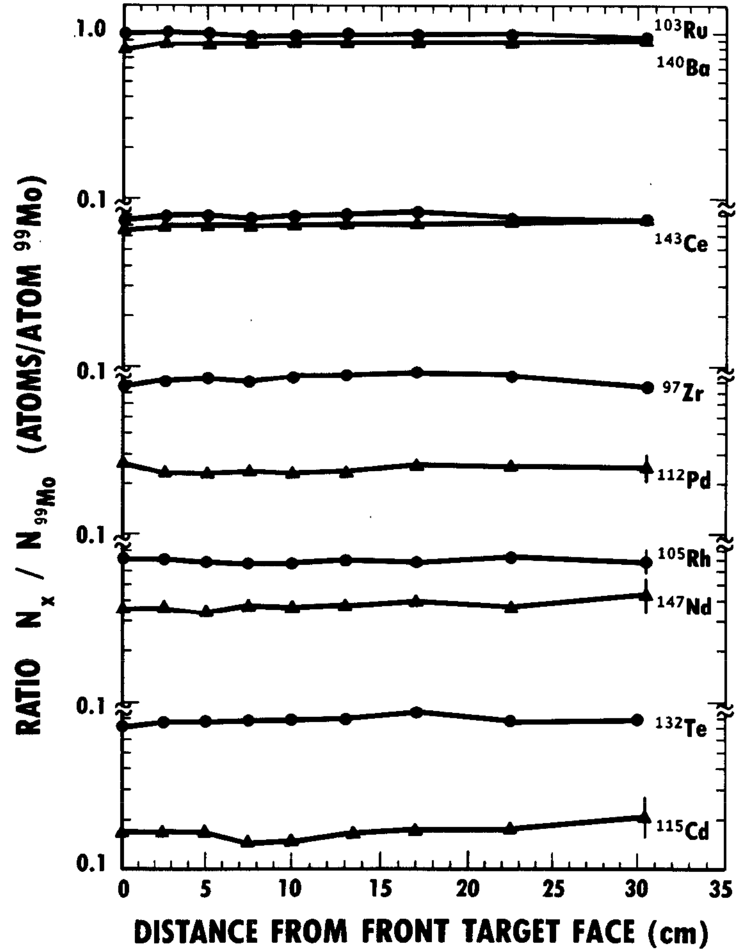

Fig. 9. Ratio of measured fission product formation to that of ^{99}Mo as a function of target axial-position.

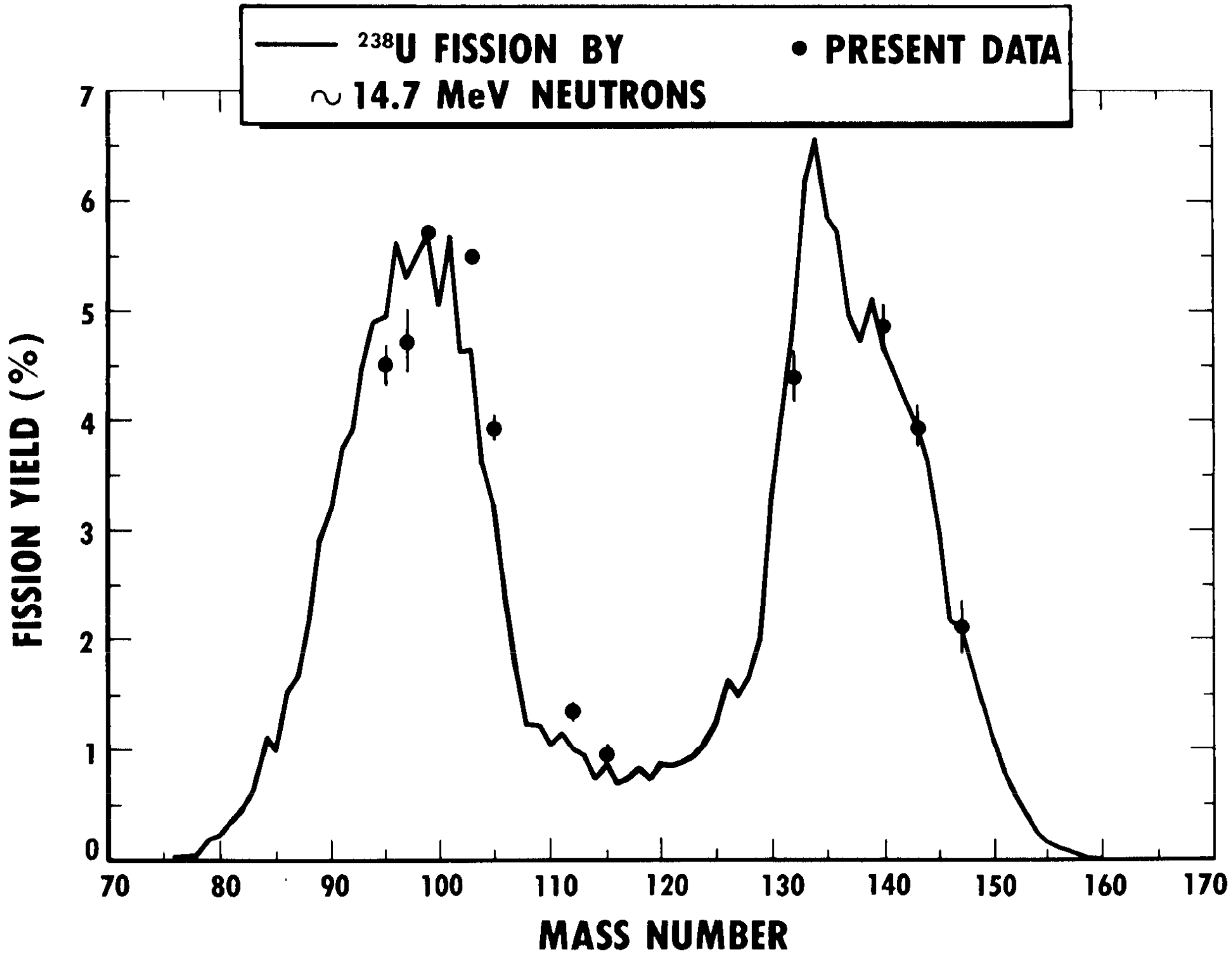

Fig. 10. Present normalized fission product yields versus mass number compared to ~14.7-MeV-neutron fission of ^{238}U (Ref. 13).

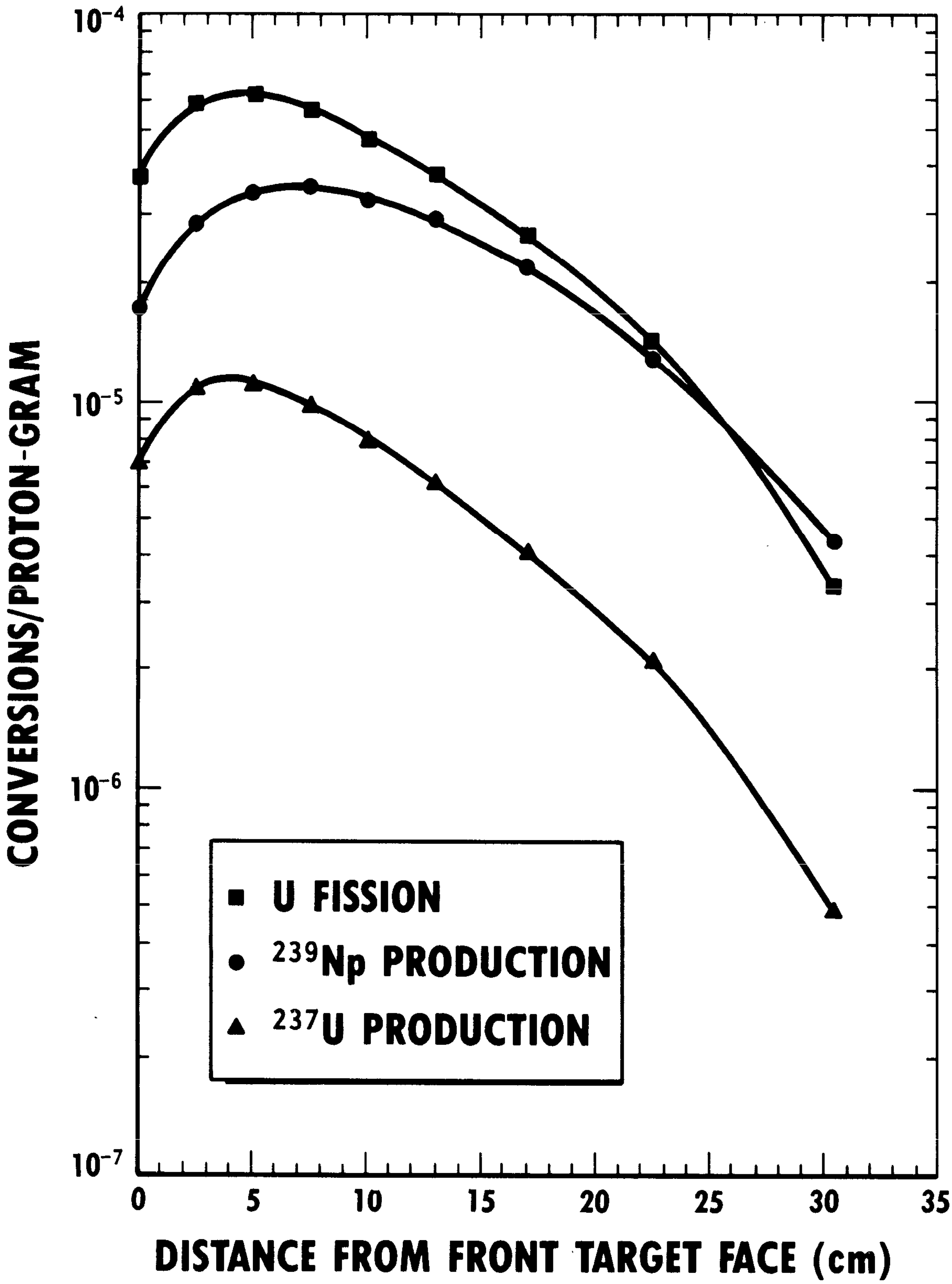

Fig. 11. Axial distribution of fissions, ^{239}Np, and ^{237}U in the depleted uranium target.