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DEVELOPMENTS IN AREAS OF ON-LINE FISSION-YIELD AND DIRECT MASS MEASUREMENTS AT THE LOS ALAMOS SCIENTIFIC LABORATORY†

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ABSTRACT

On-line studies of ^{238}U fission yields with fission spectrum neutrons were made with the Bernas surface-ionization technique at the Godiva IV burst reactor facility. The target was 300 mg of ^{238}U in a porous graphite mixture. Ion emission rates were measured by Z-direction motion of the collector. The isotope collection time was varied from 0.1 to 4.0 seconds with respect to the reactor burst. Six sets of data gave the relative yields of Cs in the mass region 138 to 146. The absolute independent fission yields were obtained by normalization using radiochemical data and chain yield estimates.

Work has also progressed toward encapsulating the Bernas source in a thin graphite container to reduce the hazards of working with highly radioactive targets. Ion emission rate measurements on encapsulated ^{235}U indicated that the ∿ 0.2 second ^{146}Cs is an accessible nuclide.

One of the authors (Leon Forman) has been charged with organizing the direct mass measurement section of the LAMPF on-line isotope separator proposal. At the Skövde meeting, we should like to report the scope of this work and the participants and their institutions.

INTRODUCTION

At the 1970 EMIS conference at Marburg, Germany, we described a method developed at Los Alamos for determining independent yields of alkali nuclides from neutron induced fission.(1) This manuscript describes changes in that technique for studies of fission induced by fission spectrum neutrons and for working with highly radioactive fissile targets.

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In addition, a program for the direct mass measurement of radioactive nuclides produced at the proposed LAMPF on-line isotope separator is briefly described. So far, this endeavor has resulted in a collaboration representing virtually all active mass measurement groups on the North American Continent and two on-line mass spectroscopy groups.

FISSION YIELD MEASUREMENTS

Briefly, the on-line fission yield measurement method at the Los Alamos Scientific Laboratory employs an intense burst of neutrons from the Godiva IV reactor to produce fissions in a target material suitably located in the source optics of the spectrograph.[1,2] The ion source design employs the Bernas surface-ionization technique.[3] Ionized alkali fission products are accelerated into a 51-cm magnetic lens and collected on the focal plane of the spectrograph for a preset time. Each mass position is later assayed by beta counting. The resulting isotopic abundance analysis can then be normalized using available radiochemical data.

The collector is designed so that it can move at a steady rate in a direction transverse to the beam spectrum. The burst signal initiates the motion, and the resulting beam deposition trace on the collector describes the ion emission from the source as a function of time.

The standard method of fabricating a target is first to extrude a mixture of uranium oxide and graphite with furfuryl alcohol as a binder, through a die and curing it at a high temperature in an inert atmosphere. The result is a stock of target material in the form of a hard, thin rod of a mixture of uranium carbide and graphite. The target is made by breaking off a suitable length from this rod.

The relative abundances of ^{138}Cs, ^{139}Cs, ^{141}Cs, ^{143}Cs, ^{145}Cs, and ^{146}Cs formed by fissioning ^{238}U with fission spectrum neutrons have been measured recently by this method. The Cs nuclides of mass 140 and 144 were not observed because the beta activity in these mass chains is too small to yield information. The target was a 3.6 mm diameter cylinder 9.5 mm long containing 300 mg of high purity ^{238}U (less than 0.025% ^{235}U) in the graphite matrix. The target was enveloped in Ta to form the source oven and maintained at a steady 1880°C during each irradiation.

For this work, the center of the reactor was moved to about 35 cm from the target with all possible moderating matter removed. A typical irradiation burst lasts for about 30 microseconds and produces a total of about 10^{17} neutrons.

The emission rate for Cs from this source oven, when maintained at 1880°C, was determined as a function of time using the moving collector technique in order to correct the observed Cs abundances for decay and ingrowth from precursors during the finite collection for the ^{238}U yield times. Collection times were varied from 0.1 to 4.0 sec in the course of six irradiations. The weighted averages of the corrected relative abundances are normalized to ^{141}Cs as unity and listed in Table I.

The estimates for the independent fission fission yields in Table I were obtained from the relative abundances by normalizing as follows: 1) The fractional cumulative yields for ^{139}Xe and ^{141}Xe have been recently measured.[4] The fractional independent yield of Cs for each mass chain was estimated by subtracting the cumulative yield of Xe from unity and correcting for the small contribution from Ba that can be inferred from systematics.[5] The fractional cesium yields thus obtained are $0.052 \begin{smallmatrix}+0.0050\\-0.0063\end{smallmatrix}$ and $0.395 \begin{smallmatrix}+0.027\\-0.075\end{smallmatrix}$ respectively. 2) These values were multiplied by the respective mass chain yields as listed by Meek and Rider[6] to provide estimates for the independent yields of ^{139}Cs and ^{141}Cs. The relative abundances are normalized to these values and given in Table I. The independent fission yields are plotted in Fig. 1 and compared to theoretical values.

The solid curve is obtained by multiplying the chain yields from Ref. 6 by the fractional independent yields of Cs from Ref. 5 and the dotted curves are ranges of probable values. The normalized yields tend to be lower. The Cs (Z = 55) yields are also lower relative to those of adjacent even Z nuclides in thermal fission of ^{235}U.[7]

TABLE I

RELATIVE AND ABSOLUTE YIELDS OF Cs FROM FISSION OF ^{238}U WITH FISSION SPECTRUM NEUTRONS

Mass	Relative Abundance	Absolute Independant Fission Yield, %
138	0.033 ± 0.024	0.057 ± 0.042
139	0.198 ± 0.034	$0.333 \begin{smallmatrix}+0.053\\-0.059\end{smallmatrix}$
141	1.00 ± 0.11	$1.69 \begin{smallmatrix}+0.27\\-0.30\end{smallmatrix}$
142	1.10 ± 0.14	$1.86 \begin{smallmatrix}+0.33\\-0.36\end{smallmatrix}$
143	1.06 ± 0.17	$1.79 \begin{smallmatrix}+0.36\\-0.39\end{smallmatrix}$
145	0.298 ± 0.034	$0.502 \begin{smallmatrix}+0.087\\-0.095\end{smallmatrix}$
146	0.065 ± 0.014	0.109 ± 0.025

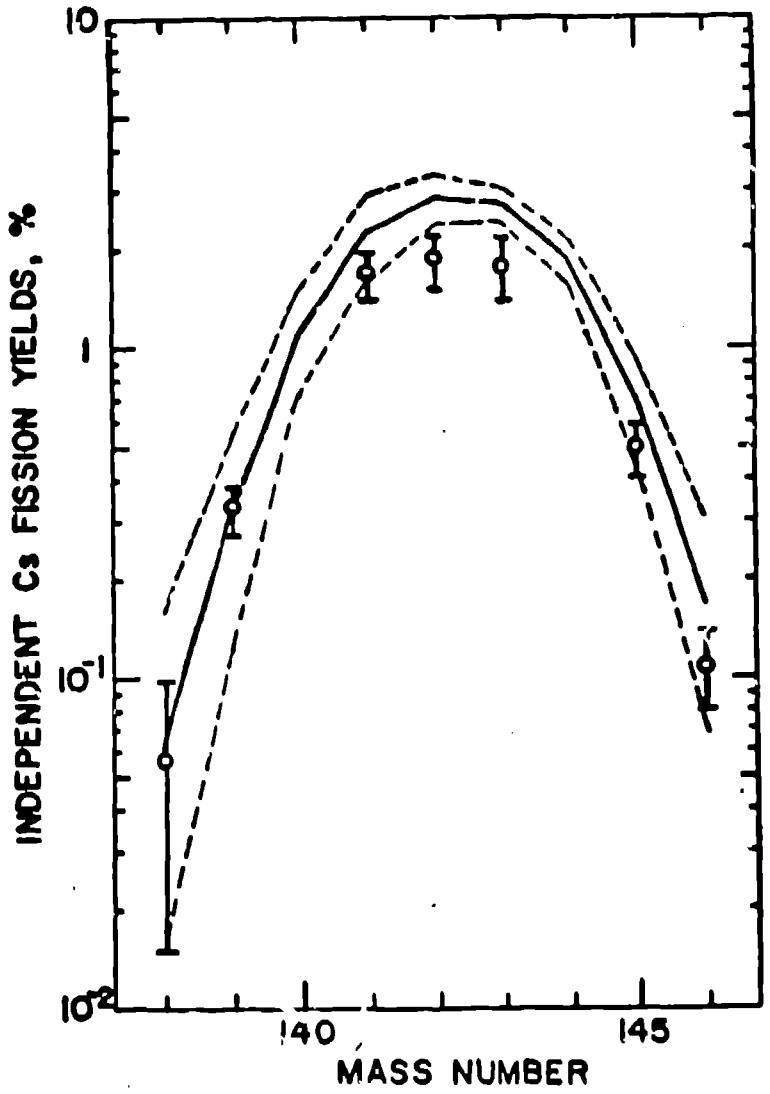

Fig. 1. Comparison of the independent fission yields of Table I (plotted as o's) compared to theoretical estimates obtained by multiplying chain yields[6] by fractional fission yields[5] (solid curve). The dashed curves indicate ranges of probable values.

ENCAPSULATION OF HIGHLY RADIOACTIVE TARGETS

A preliminary experiment has been conducted to see whether Cs atoms produced by fissioning ^{235}U can diffuse readily through the wall of a graphite capsule. The purpose of the work was to determine whether one could reduce the hazard of working with a highly radioactive target nuclide by encapsulating it and still give a satisfactory on-line source. A capsule to contain a ^{235}U target was machined as a cylindrical graphite sleeve 0.51 mm thick, closed at one end and large enough to accommodate a 6.35 mm long and 1.8 mm diameter target. After inserting the target, the capsule was sealed with a graphite stopper by brazing with nickel.

Results with and without the capsule, from an oven temperature of 1850°C indicate that the capsule reduced the ion emission only by roughly 30% for 5 sec collection times. Further study of the time dependence of the ion emission rate from the source with the capsulate target, using the moving collector technique, revealed that about 35% of the ions available are released quickly with a mean time of about 0.15 sec. In the course of calibration and testing, the source oven and capsulate target were heated repeatedly and extensively to temperatures as high as 1900°C. But on examination after the experiment, no alpha activity was detectable outside the capsule. This evidence justifies confidence for encapsulation as a means of containment.

DIRECT NUCLIDIC MASS MEASUREMENTS AT LAMPF

It is anticipated that the Clinton P. Anderson Meson Physics Facility (LAMPF) will be one of the most prolific sources of radioactive nuclides. B. J. Dropesky is presenting at this conference the status of the proposed on-line isotope separator project for LAMPF which would isolate many of these nuclides. A collaboration has been formed for the purpose of determining the nuclidic masses of these radioactive species. Coauthorship in the on-line isotope separator proposal section[8] describing these mass measurements includes: W. H. Johnson (University of Minnesota), R. C. Barber (University of Manitoba) and C. M. Stevens (Argonne National Laboratory) representing the direct mass measurement groups and L. Forman (Los Alamos Scientific Laboratory spokesman) and P. L. Reeder (Battelle Pacific Northwest Laboratories) representing on-line mass spectroscopy efforts. The broad basis for scientific interest in these mass measurements was documented by S. G. Nilsson, J. R. Nix, P. A. Seeger and W. J. Swiatecki.

Three methods are considered for the direct mass measurements. They are off-line determinations of longer-lived species, on-line studies using the isotope separator as an integral part of the mass measurement instrument, and introduction of the isotope separator beam into a high resolution double focusing instrument. All three methods have their respective advantages.

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(8) "Proposal for an On-line Mass Separator Facility at LAMPF to Study Products of Reactions Induced by an Intense 800 MeV Proton Beam", B. J. Dropesky et al., in preparation. This proposal will contain a section entitled "A Nuclidic Mass Determination Program for Radioactive Nuclides Produced at LAMPF" by R. C. Barber, L. Forman, W. H. Johnson, S. G. Nilson, J. R. Nix, P. L. Reeder, P. A. Seeger, C. M. Stevens, and W. J. Swiatecki.