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Los Alamos National Laboratory Fission Basis

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Abstract

Los Alamos has been measuring both fission spectrum and thermal fission product yields of U-235, U-238 and Pu-239 for decades. These have been reported in the literature as fission product ratios (so-called R-values) and from these data we can construct ratios of fission product yields in different neutron environments (so-called Q-values). In the 1970's we participated in the Inter-laboratory Liquid Metal Fast Breeder Reactor (LMFBR) Reaction Rate (ILRR) collaboration on measurements at both the BIG TEN critical assembly and the Omega West Reactor. Although these experiments were conducted decades ago, it is only now that we document the results in great detail together with detailed uncertainty estimates, and translate the results into fission product yields (FPY) so that they can be compared with other literature values. These fission product data agree with measurements by Maeck and other authors using mass-spectrometry methods, and other data within the ILRR collaboration using gamma spectroscopy for quantifying fission products. We note that the measurements contradicted earlier 1950s historical Los Alamos estimates by 5-7%, most likely due to self-shielding corrections not made in the early thermal measurements. Our experimental results provide a confirmation of the England-Rider ENDF/B-VI evaluated fission-spectrum fission product yields that were carried over to the ENDF/B-VII.0 library, except for ^{99}Mo where the present results are about 3% relative higher for neutrons incident on ^{239}Pu and ^{235}U . Additionally, our results illustrate the importance of representing the incident energy dependence of fission product yields over the fast neutron energy range for high-accuracy work, for example the ^{147}Nd from neutron reactions on plutonium. An upgrade to the ENDF library, for ENDF/B-VII.1, based on these and other data will be released in December 2011.

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Take Away

- **Re-analysis of LANL radiochemical fission basis**
 - Consistent with past analysis
 - Detailed uncertainty analysis
 - Published full results
- **Resolved long standing discrepancy for ^{239}Pu FPY, especially ^{147}Nd**
 - ENDF/B-VI was largely correct
 - New results integrated into ENDF/B-VII (December 2011)
 - Including energy dependence
 - LLNL, JEFF, JENDL, and other libraries will likely need some modification as well

Outline

- The Problems
- History
- Dosimetry Experiment
- Energy Dependence (^{147}Nd)
- Outlook

Two Main Problems Motivated This Work (Early 2000's)

- **Different laboratories were calculating total fissions for the same plutonium sample material and getting different results.**
 - Some cases up to 7-8% different
- **Nuclear simulation codes have become very precise and are now limited by the errors in the nuclear data.**
- **Understand the errors!**
 - The published literature (and therefore the libraries (ENDF, JENDL, JEFF, etc.)) contained data from early measurements that were not corrected for self-absorption
 - LANL had participated in the ILRR program in the 1970's and discovered this issue
 - Notes in internal publications and ILRR program documents, but no reports were made in widely distributed publications
 - Several years of research, getting original logbooks, gathering all the information from these critical experiments was undertaken to publish these results and update the libraries (ENDF-VII to be released in December).

LMFBR – Liquid Metal Fast Breeder Reactor

- Lots of research 60's-70's
- ILRR Formed – Supported by the AEC (DOE)
 - Inter-laboratory LMFBR Reaction Rate Program
 - *Established to develop the capability to measure neutron-induced reaction rates for LMFBR fuels and materials development program*
 - ^{235}U , ^{238}U and ^{239}Pu - 2.5% accuracy for (n,f)
- Develop tools to quickly and accurately measure burn-up (total fissions) - Specific Gamma Spectroscopy
- LANL - Using Separation Chemistry & Counting
- Facilities
 - EBR-II - Experimental Breeder Reactor II (INL)
 - CFRMF – Coupled Fast Reactivity Measurement Facility (INL)
 - BIGTEN Critical Assembly (LANL)
 - Omega West Reactor (LANL)
 - $\Sigma\Sigma$ -Mol – Natural Uranium Secondary Standard Neutron Field (Belgium)

Experimental Breeder Reactor II

Aerojet Nuclear Company (ANC)
Argonne National Laboratory (ANL)
Atlantic Richfield Hanford Company (ARHCO)
Atomics International (AI)
Brookhaven National Laboratory (BNL)
Joint Center for Graduate Study (JCGS)*
Hanford Engineering Development Laboratory (HEDL)
Lawrence Livermore Laboratory (LLL)
Los Alamos Scientific Laboratory (LASL)
National Bureau of Standards (NBS)**
Oak Ridge National Laboratory (ORNL)
University of California at Santa Barbara (UCSB)

Two Ways to Calculate Total Fissions

$$F = A_i K_i = N_i / Y_i$$

Materials to be Irradiated

- **Must have well characterized materials**
- **Reference Foils (Electroplated Materials)**

- Traced with ^{232}U
- Comparative Fission Counting
- Mass known to about 1.5%

Element/ isotope	Isotope atom %
$^{235}\text{U}_3\text{O}_8$	
^{233}U	<0.0001
^{234}U	0.029
^{235}U	99.912
^{236}U	0.0165
^{238}U	0.0414

- **Target Foils**

- Direct Weight
- IDMS – Isotope Dilution Mass Spectrometry
- Alpha Counting
- Comparative Fission Counting
- Mass known to less than 2%

Material	Mass (mg)
^{235}U (expt 2725)	$0.5085 \pm 1.42\%$
^{235}U (expt 2755)	$0.5663 \pm 0.87\%$
^{239}Pu (expt 2726)	$0.3748 \pm 1.53\%$

• Cold-rolled, polished (uranium), weighed, and sealed in inert atmosphere

• Prepared by Rocky Flats

• Cold welded into packets, weighed by ORNL

Irradiation Facilities

- **Omega West Reactor**
 - Light Water Reactor
 - Enriched Uranium Fuel
 - 8 MW Nominal Power
- **Graphite Thermal Column**
 - Cd Ratio = 5,000-10,000
 - 5×10^{10} n/cm²-s

- **BIGTEN Critical Assembly**
 - Average 10% Enriched ²³⁵U
 - 6" Depleted Uranium Reflector
 - 8×10^{10} n/cm²-s

Fission Counting

- **Material Foils Inside Chamber**

- **Insert Fission Chamber into Irradiation Port**

- Both OWR & BIGTEN able to use the same Fission Chamber

- **Uncertainties**

- Dead Time, Deposit Mass, Minor Isotope Fissions, Non-Counted Fragments
- Total Uncertainty ~ 1.5-2.0%

- **Reference Foil**

- Fissions/mg
- $3.144 \times 10^{11} (\pm 1.7\%)$

- **NBS (NIST) Designed Fission Chamber**

- Low Activation Background
- No Hydrogenous Material
- Calibrated with known ^{252}Cf source

Chemistry

- **Dissolution Chemistry**
 - Foils dissolved in mineral acids
 - Aliquoted and Diluted
- **Separation Chemistry**
 - Specific Chemistry Procedures – LA-1721
 - Multiple Preparations for each nuclide
 - Multiple Chemists for certain nuclides
- **Plate/Stipple Material on Plates for counting**

Counting

- Cumulative chemistry and thickness($\pm 0.6-1.5\%$)
- Counter drift $\sim 1\%$ over 30 years
- Identical counters (Ctr X/Ctr 6 = {0.99 to 1.02})
- Daily standard counts using ^{234}Th and ^{234}Pa betas from ^{238}U source

^{147}Nd Fission Product Yields

^{147}Nd Energy Dependence

Outlook for Fission Measurements

- **Reviewing unpublished critical assembly and reactor data**
 - Publish results
 - Van De Graff accelerator (High Energy Neutrons 1-24 MeV) (LANL)
- **New Experimental Efforts**
 - Further reduce uncertainties to less than 1%
 - LLNL developing TPC (Time Projection Chamber) (LANSCE)
 - Measure neutron output spectra in energy regions that have not been measured
 - LANL developing ChiNu – Neutron Detector Array (LANSCE)
- **New Facilities**
 - CEF – Critical Experiments Facility (NNSS)
 - Planet, Comet, Flattop, Godiva
 - Goal of running an experiment with Planet this year
- **Updating ENDF**
 - Including “new” data, and energy dependencies
 - ENDF/B-VII should be available this December

Van De Graff

Godiva

Available online at www.sciencedirect.com

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**Nuclear Data
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Fission Product Data Measured at Los Alamos for Fission Spectrum and Thermal Neutrons on ^{239}Pu , ^{235}U , ^{238}U

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R. Prestwood, V. Armijo, J. Hasty, et al, 1945-present.

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Sheets**

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Fission Product Yields from Fission Spectrum $n+^{239}\text{Pu}$ for ENDF/B-VII.1

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