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STATUS OF FISSION YIELD MEASUREMENTS

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# 1. INTRODUCTION

Fission yield data, for all neutron energies, play an important role in many of the disciplines (fuel burnup, fission rate measurements, dosimetry, reactor physics, damage studies, post irradiation fuel analysis, etc.) being discussed at this symposium. This paper reviews and presents a brief discussion of the current status of fission yield measurement programs being conducted in the major laboratories throughout the world, and, when possible, to identify future activities. The contents of this paper are based on two sources: 1) a mailing by the author to various principal investigators furnishing a form which could be used to describe their most current work, and 2) reference to the most recent IAEA Nuclear Data Section document "Progress in Fission Product Nuclear Data"<sup>1</sup>, which is a compilation of current measurement activities relative to fission product nuclear data.

~~The general organization of the paper is by fissioning nuclide and the neutron energy region being examined.~~

## ABSTRACT

*and yield compilation*  
Fission yield measurement activities in the major laboratories of the world are reviewed. In addition to a general review of such activities, a brief summary of yield measurement activities ~~is presented~~ by fissioning nuclide is presented. A new set of fission product nuclear data being conducted in the U.S. is ~~presented~~.

## 2. GENERAL REVIEW

### 2.1 Fission Yield Measurements

The principal laboratories currently engaged in more extensive fission yield measurements are located in France, Germany, India, Japan, Switzerland, United Kingdom, and USA. The most ambitious programs appear to be those being conducted in the USA.

Perhaps the most extensive absolute yield measurement program is the long-term program being conducted at the Idaho National Engineering Laboratory (INEL) by Maeck and co-workers. In this program, isotope dilution mass spectrometry is used to measure thermal and fast reactor chain yields for over 40 stable and long-lived fission products. In the past few years, absolute fission yields have been reported for  $^{233}\text{U}$ ,  $^{235}\text{U}$ , and  $^{238}\text{U}$  fast fission<sup>2</sup>,  $^{239}\text{Pu}$  and  $^{241}\text{Pu}$  fast fission<sup>3</sup>,  $^{240}\text{Pu}$  and  $^{242}\text{Pu}$  fast fission<sup>4</sup>,  $^{237}\text{Np}$  fast fission<sup>5</sup>, and recently  $^{235}\text{U}$  and  $^{239}\text{Pu}$  thermal fission<sup>6,7</sup>. In general, the uncertainties associated with the measured yields are in the 1-2% range. Currently, <sup>this laboratory is measuring yields</sup> yield measurements are being made <sup>for</sup> on samples of  $^{233}\text{U}$ ,  $^{235}\text{U}$ ,  $^{239}\text{Pu}$ , and  $^{241}\text{Pu}$  which <sup>were</sup> had irradiated in a harder spectrum of Experimental Breeder Reactor -II (EBR-II), than those previously reported. <sup>2,3,4,5 Fission yield</sup> A new irradiation and measurement program <sup>sample irradiated in</sup> for the Fast Flux Test Facility (FFTF) is discussed in detail in Section 4. ←

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A <sup>major</sup> companion effort <sup>associated with</sup> related to the INEL program is the correlation of reactor fission yields with neutron energy<sup>8,9</sup>.

Glendenin, Gindler, and co-workers of the Argonne National Laboratory (ANL) are currently conducting a systematic study of fission yields versus neutron energy for mono-energetic neutrons in the range of 0.1 to 8.0 MeV, using the ANL fast neutron generator. The measurement techniques are direct high resolution gamma ray spectrometry and radiochemical separations. In general, the uncertainties range from 3-15%, depending on the magnitude of the yield and the measurement

technique. To date, mono-energetic yields have been published for  $^{238}\text{U}$ <sup>10,11</sup> and preliminary results have been reported for  $^{232}\text{Th}$ <sup>12</sup>. Similar experiments are in progress for  $^{233}\text{U}$ ,  $^{235}\text{U}$ , and  $^{239}\text{Pu}$ <sup>14</sup>.

Nethaway and co-workers at the Lawrence Livermore Laboratory (LLL) have recently reported <sup>yields for  $^{240}\text{Pu}$  irradiated with neutrons<sup>13,14</sup></sup>  $^{240}\text{Pu}$  yields for 14.8 MeV fission<sup>13</sup> and a "slightly degraded" fission spectrum<sup>14</sup>. <sup>Fission yields for</sup> At this program 75 fission products were measured in the 14.8 MeV irradiation, and 45 fission products in the critical assembly experiment. The measurement technique was direct high resolution gamma ray spectrometry. The average uncertainty associated with the reported yields is estimated to be about 5% relative. Similar experiments have been completed for  $^{241}\text{Am}$ , but no values have yet been reported, ~~for each target nuclide,~~ a limited number of independent and cumulative fractional yields<sup>also</sup> are reported<sup>13,14</sup>.

At Oak Ridge National Laboratory (ORNL), Dickans and co-workers have recently extracted fission yield data from an experiment designed to measure the total beta and gamma energy release <sup>as</sup> or a function of time for the short-term thermal fission of  $^{235}\text{U}$ ,  $^{239}\text{Pu}$ , and  $^{241}\text{Pu}$ . Using a large volume Ge(Li) detectors, the yields for 16 and 36 mass chains were measured for  $^{241}\text{Pu}$  and  $^{239}\text{Pu}$  thermal fission, respectively. The absolute uncertainties range from 3 to 25%, relative. The  $^{241}\text{Pu}$  results have been published<sup>15</sup>, and the  $^{239}\text{Pu}$  data are being prepared for publication.

<sup>During</sup> At the Second ATSM-EURATOM <sup>Symposium on Reactor Dosimetry,</sup> Conference several laboratories associated with <sup>in the US</sup> an interlaboratory reaction rate measurement program, presented papers<sup>16,17</sup> giving evaluated consensus fission yield data for those fission product isotopes of primary interest to fission rate and dosimetry measurements. In this cooperative experiment, the measurement method was high resolution gamma spectrometry and the number of fissions was established using a fission chamber. It is anticipated that a status report relative to this program will be presented in the current symposium.

At the Battelle Pacific Northwest Laboratories (PNL), Ballou and Reeder and Co-workers have measured the independent and cumulative  $^{235}\text{U}$  thermal fission yields of some very neutron-rich nuclides using the "on-line" mass spectrometer, SOLAR (Spectrometer for On-Line Analysis of Radionuclides)<sup>18</sup>. This is an on-line mass spectrometer which incorporates a  $^{235}\text{U}$  target in a surface ionization source located in the thermal column of a ~~TRIGA~~ <sup>1 MW</sup> TRIGA reactor at Washington State University, Pullman, Washington, USA. In this instrument, a short burst of neutrons from the TRIGA reactor is used to produce various isomers of Br, Rb, In, I, and <sup>Ca</sup> ~~Cs~~ fission products within the surface ionization source. Selective ionization performs the rapid chemical separation to give the desired nuclides as beams of ions. The ions are collected on a moving tape collector system for a short time-interval during and after the neutron pulse. Beta and gamma counting are used to follow the decay of the isomers to determine the yield of each. A knowledge of the particular decay schemes is the limiting factor associated with accuracy of the results. More recent studies are directed on the characterization of individual delayed neutron emitters.

Using a 1.5 Mw TRIGA reactor and the fission-fragment recoil mass spectrometer, HIAWATHA<sup>19</sup>, Wehring and co-workers at the University of Illinois, have measured and reported <sup>20,21</sup>  $^{235}\text{U}$  thermal fission yields. The technique is reported to produce yields with associated uncertainties of ~1% relative for the largest mass yields. A unique capability of this technique is that chain and independent yield data can be obtained for all nuclides <sup>having</sup> independent yields greater than 0.1%.

HIAWATHA, consisting of a cylindrical focusing electrostatic analyzer and time-of-flight <sup>no TP</sup> system, is used to determine fragment masses while fragment energy loss is used to identify fragment atomic numbers in a multiparameter experiment. All fragment velocities and charge states are covered in this direct physical measurement of fission yields. Mass resolution of 0.5 amu and atomic number resolution of about 1-2 has been achieved. Uncertainties about 1% have been achieved for the largest mass yields. New work planned for this instrument is the

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investigation of the dependence of prompt neutron yields on fission fragment mass, atomic number, and energy.

At the Los Alamos Scientific Laboratory (LASL), recent efforts have centered <sup>on</sup> ~~about~~ the measurement of the relative fission yields of various krypton and xenon isotopes for thermal, degraded-fission-spectrum, and 14-MeV neutron-induced fission of  $^{235}\text{U}$  and  $^{239}\text{Pu}$ , and for degraded-fission-spectrum and 14-MeV fission of  $^{236}\text{U}$  and  $^{238}\text{U}$ <sup>42</sup>. Wolfsberg at LASL, in cooperation with Meixler and Denschlag at the University of Maine, have recently reported <sup>43</sup> fractional cumulative yields of <sup>for several</sup> ~~of~~ <sup>short-lived</sup> krypton and xenon isotopes for the thermal fission of  $^{249}\text{Cf}$  and spontaneous fission of  $^{250}\text{Cf}$ .

<sup>In</sup> At the United Kingdom, the principal centers for fission yield measurements are at Dounreay and Harwell. The Dounreay program, under the direction of W. Davis and V. M. Sinclair, is primarily directed to the measurement of the fast reactor yields of the stable isotopes of neodymium and of  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$ , and  $^{144}\text{Ce}$ ; isotopes of interest in the measurement of nuclear fuel burnup. In this program, samples of enriched  $^{235}\text{U}$ ,  $^{238}\text{U}$ ,  $^{239}\text{Pu}$ , and  $^{241}\text{Pu}$  are being irradiated in the Prototype Fast Reactor (PFR) at Dounreay. ~~It is expected that~~ The irradiation will be completed by the end of 1979. For  $^{235}\text{U}$ ,  $^{239}\text{Pu}$ , and  $^{241}\text{Pu}$ , the atom percent fission is expected to exceed 15%, and for  $^{238}\text{Pu}$  and  $^{240}\text{Pu}$ , 1 to 4%. The principal measurement technique will be isotope dilution mass spectrometry. The number of fission will be determined using the heavy element difference method. Expected uncertainties are 2% relative for the fission yields for  $^{235}\text{U}$  and  $^{239}\text{Pu}$  and 6% for  $^{238}\text{U}$ ,  $^{240}\text{Pu}$ , and  $^{241}\text{Pu}$ . Final results should start <sup>b</sup> becoming available by late 1980<sup>22</sup>. At the present time, no new fission yield experiments are planned.

At Harwell, Cuninghame and Willis have irradiated mg samples of  $^{235}\text{U}$ ,  $^{238}\text{U}$ ,  $^{239}\text{Pu}$  in the core and blanket region of the fast reactor facility, ZEBRA, to measure the effect of the change of reactor neutron spectrum on fission yields. The measurement technique will involve direct high resolution gamma spectrometry and radiochemical separations. The number of fissions are determined <sup>using</sup> ~~from~~

fission chamber. About 20 nuclides will be measured with uncertainties expected to range from 5 to 10%, relative. No results have been published to date. It is estimated that 6 to 12 months will be required to complete the work.<sup>23</sup>

Also at Harwell, McKean and Crouch<sup>24</sup> are conducting experiments to determine the fission yield of <sup>3</sup>H<sub>A</sub> (tritium) for several isotopes of uranium and plutonium in both thermal and fast reactor spectra. Samples of <sup>235</sup>U and <sup>239</sup>Pu in solution have been irradiated in a thermal flux, and metal and oxide samples of enriched <sup>235</sup>U, <sup>239</sup>Pu, <sup>240</sup>Pu, <sup>241</sup>Pu, and <sup>242</sup>Pu<sub>A</sub> are being irradiated in the fast reactor facility, ZEBRA. The fission produced tritium is converted to <sup>3</sup>H<sub>2</sub>O, separated from other fission products, and measured using liquid scintillation counting. The expected accuracy is 10% relative.<sup>24</sup>

The fission yield measurement program in Switzerland is continuing under the direction of H. R. Von Guenten of the Eidg. Insitute für Reaktorforschung at Würenlingen and the Institute für anorganische, analytische and physikalische Chemie at the Universität Bern. Presently<sup>Currentlly</sup>, the work is concentrated in three areas. ~~The first area is~~ First-is-work related to the measurement of yields of <sup>248</sup>Pm and selected Nb isotopes for <sup>233</sup>U, <sup>235</sup>U, and <sup>239</sup>Pu thermal fission. The second area<sup>is</sup> is the measurement of <sup>239</sup>Pu fission yields for the 0.3eV fission resonance. The third area, which involves a new program, is the measurement of the absolute chain yields of <sup>232</sup>Th in a fast reactor spectrum. References 25, 26, and 27 identify recent publications.<sup>46</sup>

At the Institute of Atomic Energy, Kyoto University in Japan, the principal effort<sup>has</sup> have been on the measurement of independent and cumulative fission yields for various isotopes of tin, antimony, tellurium, iodine, xenon, cesium, rubidium, and promethium for <sup>233</sup>U, <sup>235</sup>U, and <sup>239</sup>Pu<sub>A</sub>. This work is conducted using the 5MW<sub>or</sub> research reactor at Kyoto University and radiochemical separations and high resolution gamma<sup>ray</sup> spectrometry. Recent studies involve measurements of the independent yields for <sup>138</sup>I, <sup>90</sup>Rb, and <sup>148</sup>Pm for <sup>233</sup>U, <sup>235</sup>U, and <sup>239</sup>Pu thermal fission. The work on <sup>138</sup>cp has been finished and work on the other two

isometric yields is expected to be completed by the end of 1979.<sup>30</sup>

In Germany, the principal laboratories involved in fission yield measurements are located at Mainz and Karlsruhe. Denschlay and co-workers, at the Institut für Kernchemie, Universität Mainz, are conducting an extensive program relative to the measurement of independent, fractional independent, and fractional cumulative yields for many isotopes for  $^{235}\text{U}$  thermal fission. The facilities involve a TRIGA reactor and the ~~radiochemical techniques involve~~ automated rapid radiochemical separations followed by high resolution gamma spectrometry. Recent activities have involved measurement for the short lived fission products of <sup>yttrium,</sup> yttrium, niobium, zirconium, molybdenum, barium, lanthanum, cerium, and gallium for  $^{235}\text{U}$  thermal fission. New activities involve the measurement of the fractional cumulative yields for various short-lived krypton and xenon isotopes for  $^{232}\text{Th}$  fast fission<sup>31</sup>. To date, most of the results have been published in the Institutes' Annual reports.

In a cooperative GEA-France Euratom program, termed TACO (Taux de Combustion), Koch at the Transuranium Institute at Karlsruhe, has been involved in the measurement of fast reactor fission yields for several heavy nuclides. In this experiment separate encapsulated samples of  $^{232}\text{Th}$ ,  $^{233}\text{U}$ ,  $^{235}\text{U}$ ,  $^{236}\text{U}$ ,  $^{238}\text{U}$ ,  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$ ,  $^{241}\text{Pu}$ ,  $^{242}\text{Pu}$ ,  $^{237}\text{Np}$ ,  $^{241}\text{Am}$ , and  $^{243}\text{Am}$  <sup>contained in a</sup> were inserted into a standard fuel rod and were irradiated in the French fast reactor, RAPSODIE, to an integrated neutron fluence of  $\sim 10^{23} \text{ n/cm}^2$ . The measurement techniques involved direct gamma ray spectrometry for  $^{125}\text{Sb}$ ,  $^{137}\text{Cs}$ , and  $^{144}\text{Ce}$ , and isotope dilution mass spectrometry for <sup>the isotopes of</sup> Cs, Nd, Sm, Eu, and Gd. Approximately 70% of the heavy mass peak was measured and the balance estimated to give the total number of fissions<sup>32</sup>. Fast reactor yields for  $^{235}\text{U}$ ,  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$ ,  $^{241}\text{Pu}$ , and  $^{242}\text{Pu}$  have been reported<sup>33</sup>.

In 1978, Debertine<sup>34</sup>, Physikalisch-Technische Bundesanstalt, Braunschweig, reported fast neutron fission yields for  $^{238}\text{U}$ . In this experiment, the neutron source was a  $^{252}\text{Cf}$  source mounted in air 1-cm away from the  $^{238}\text{U}$  targets. The measurement techniques was high-resolution gamma ray spectrometry. The authors reported uncertainty associated with the measured yields is 2-5%, relative.



in French

Several fission yield programs are in progress in France at various nuclear research centers. Laurec and Adam at Bruyeres-Le-Chatel are measuring the cumulative yields of many radioactive fission products for the fissioning of  $^{233}\text{U}$ ,  $^{235}\text{U}$ ,  $^{238}\text{U}$ , and  $^{239}\text{Pu}$  with fission spectrum and 14.7 MeV neutrons. The number of fissions is <sup>determined</sup> measured using a fission chamber, and the fission product activities are measured directly using a calibrated high resolution gamma ray spectrometer. The mass of the target nuclide is determined either by alpha or mass spectrometry. This work should be completed by the end of 1979 <sup>and</sup> at which time a CEA report will be issued.

Blachot and co-workers at Grenoble are measuring several independent and fractional cumulative yields for  $^{235}\text{U}$  thermal fission and 3 MeV fission using the Melusine reactor and a 3 MeV <sup>V</sup> neutron generator. The principal measurement technique is high resolution gamma ray spectrometry, and the estimated uncertainty in the measured values is 5-10% relative. Preliminary data have been presented <sup>in values</sup>  $^{35}$ . New work at Grenoble is the measurement of independent, fractional cumulative, and chain yields for the 3 MeV fission of  $^{232}\text{Th}$   $^{36}$ .

It is anticipated that recent French work on fast reactor fission yields using mass spectrometric techniques will be presented in this symposium.

In India, at the Bhabha Atomic Research Center at Trombay, Bombay, three different techniques are being studied and used for the measurements of fission yields. The first, involves the measurement of short-lived fission products using high resolution gamma ray spectrometry and the track etch technique with mica <sup>tracks</sup> to obtain the number of fissions. To date, work has been completed for the thermal fission of  $^{235}\text{U}$  and  $^{239}\text{Pu}$   $^{37}$ , and work is in progress for the thermal fission of  $^{233}\text{U}$  and fast fission of  $^{232}\text{Th}$ .

<sup>The second program is related to the measurement of  $^{232}\text{U}$  yields. In this work</sup>  
A For  $^{232}\text{U}$ , the yields were measured relative to  $^{235}\text{U}$  using  $^{99}\text{Mo}$  as the reference standard <sup>and</sup>. In this work catcher foils were used which were either counted directly <sup>or</sup> using high resolution gamma ray spectrometry or subject to a

radiochemical separation. This work is to be published in the near future.

A more recent program <sup>third</sup> is based on <sup>involves</sup> mass spectrometric measurements. In this program, the thermal fission yields for <sup>relative</sup>  $^{233}\text{U}$ ,  $^{235}\text{U}$ ,  $^{239}\text{Pu}$ , and  $^{241}\text{Pu}$  are being determined for about 20 mass numbers using mass spectrometric techniques, ~~for the determination of relative yields.~~

## 2.2. Compilations

There are two major fission yield compilations which are in a continuing effort. One is the work of E. A. Crouch<sup>38</sup> at Harwell. In this work, Crouch is attempting to compile and evaluate neutron induced fission product yields for all fissionable nuclides at neutron energies up to 15 MeV. In this work, ~~no~~ <sup>being</sup> results prior to 1950 are considered. Literature reported chain yields are adjusted to force agreement with the conservation laws. The goal is to generate a consistent set of values. When necessary, the evaluated yields are augmented by interpolation to fill in missing values. This data file currently exists on magnetic tape or punched cards in ENDF/B-IV format.

The second major yield data file is in the U.S. Evaluated Nuclear Data File (ENDF) for yields. This compilation is primarily the work of B. F. Rider and the ENDF yield sub-committee. At the present time Rider<sup>39</sup> has incorporated yield data for 30 fissioning systems in his compilation. This is based on approximately 18,000 entries from 1200 references. This compilation is under constant revision and is updated annually. Numerous tests of the data are made by General Electric Company, Los Alamos Scientific Laboratory, Hanford Engineering Laboratory and the University of Oklahoma prior to incorporating the data into the most recent U.S. file, ENDF/B-V. Reference 40 gives a review and discussion of the general approach used.

One consoling feature of these two compilations is that the authors are making an attempt to review all of the input data and to reconcile differences. The major difference between the two compilations is the methodology and restrictions

used in assessing errors to the recommended values. As a measure of fission yield data, ~~it is my personal feeling~~ <sup>I feel</sup> that the uncertainty restrictions imposed by Crouch on reported experimental data is too harsh and that the error analysis method used by Rider produces overly optimistic uncertainties. Although many laboratories, including our own, report yields with better than 1% relative

uncertainties, <sup>there</sup> I still believe that there are certain unknown systematic errors, <sup>which</sup> have not been included in the propagation. To be conservative, I suggest that a lower limit of 1% is a reasonable relative uncertainty to place on any yield value. For values quoted as "fast reactor fission yields" this limit may have to be increased.

<sup>Currently</sup> ~~At the present time~~, both compilations are using a three group energy basis for the data; thermal, fast, and high energy of 14.8 MeV. <sup>I</sup> ~~It is agreed~~ that all thermal data should be internally consistent and that the data can be pooled to give a best estimate of the <sup>three</sup> value. For fast reactor yields, however, the same statistical treatment cannot be used because the yields are <sup>changing</sup> with neutron energy. Very significant differences are known to exist for the isotopic composition of certain fission products just within the core of a single fast reactor<sup>8,9</sup>. This being the case, <sup>and</sup> that fast yield compilations involve data from many different fast neutron irradiations, I do not believe that uncertainties of 1% or less can be assigned to any yield value without identifying the neutron spectrum. Until a system is developed which gives yield data as a function of energy, caution must be exercised in the assignment of errors to "fast reactor fission yields". Several approaches to clarifying this problem are being explored in our laboratory.

Neutron Physics

### 3. MEASUREMENT PROGRAMS IN PROGRESS BY FISSIONING NUCLIDE

In this section, recent and current fission yield measurement <sup>activities</sup> programs are compiled according to the fissioning nuclide. When possible, the programs will be identified with neutron energy (i.e., thermal, fast reactor, mono energetic, 14.8 MeV, and others).

#### 3.1 Thorium - 232

With ~~not~~ more consideration being given to alternate fuel cycles, the number of laboratories involved in the measurement of <sup>232</sup>Th <sup>yields</sup> has increased in the past few years.

3.1.1 Fast Reactor. Essentially all measurement programs for <sup>232</sup>Th yields involve measurements using high resolution gamma ray spectrometry. Absolute <sup>232</sup>Th yields are being measured by Von Gunten in Switzerland and at the Bhabha Atomic Research Center in India. Richardson has reported yields for <sup>232</sup>Th irradiated by degraded fission spectrum neutron and measured using high resolution gamma ray spectrometry<sup>41</sup>. In all cases, the accuracy of the data will probably range from 3-10% depending upon the isotope being measured. This error range is about the same which exists for compiled data<sup>38,39</sup>.

No extensive fast reactor fission yields for <sup>232</sup>Th based on isotope dilution <sup>mass spectrometry</sup> measurements have been reported to date. Admittedly, this is a difficult, but not impossible task. With a designed experiment it should be possible to produce <sup>232</sup>Th yield data accurate to within a few percent, which would go far toward reducing existing uncertainties. Because of the low fission cross section for <sup>232</sup>Th, several grams of material must be irradiated for a relatively long time compared to <sup>235</sup>U or <sup>239</sup>Pu ( $10^{18}$  fissions is a reasonable value). Such conditions will produce significant quantities of <sup>233</sup>U which will also fission. To reliably correct for the contribution from <sup>233</sup>U fast fission, separate samples of <sup>233</sup>U

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should be irradiated under the same conditions in the same neutron spectrum. Thus, even if 10-20% of the total fissions in the  $^{232}\text{Th}$  target result from  $^{233}\text{U}$  fission, reliable corrections could be made.

Danschlag at Mainz is in the process of measuring fractional cumulative yields for short-lived krypton and xenon isotopes for fast-reactor fissioned  $^{232}\text{Th}$ .

3.1.2 Monoenergetic Neutrons. Using a fast neutron generator at ANL, Gindler and Glendenin are conducting a systematic study of  $^{232}\text{Th}$  yields as a function of neutron energy for the energy range 0.1 to 8 MeV. The measurement technique is high resolution gamma ray spectrometry. Preliminary results have been presented<sup>12</sup>. For the major fission product yields, the uncertainties are estimated at 3-5% and for the minor isotopes (<1% FY), 5-20%. Blachok at Genoble is starting to measure  $^{232}\text{Th}$  independent, fractional cumulative, and chain yields for 3 MeV neutron fission using a neutron generator and gamma ray spectrometry.<sup>36</sup>

### 3.2 Uranium - 232

Manohar and co-workers recently completed the radiochemical measurement of the fission yields for  $^{232}\text{U}$  fission. The yields were measured relative to the yield for  $^{99}\text{Mo}$  for  $^{235}\text{U}$  fission<sup>1,45</sup>.

### 3.3 Uranium - 233

3.3.1 Thermal Neutrons. One major measurement program is in sight for  $^{233}\text{U}$  thermal fission is that proposed by Wehring using the fission-fragment recoil mass spectrometer HIAWATHA<sup>19,20</sup>. The exact status of this work is unknown. Some independent and fractional cumulative yields for  $^{233}\text{U}$  thermal fission have recently been reported by Nishi<sup>29,30</sup> and Kaiser and Von Gunten<sup>25</sup>. Relative yields for about 20 mass numbers for  $^{233}\text{U}$  thermal fission are being measured, employing mass spectrometric techniques, by Chitambar and co-workers in India<sup>1</sup>. Also in

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India,  $^{233}\text{U}$  thermal yields are being measured for several short-lived fission products<sup>1</sup>.

3.3.2 Fast Reactor Neutrons. The last major work on  $^{233}\text{U}$  fast yields was reported by Maech<sup>K</sup> and co-workers for samples irradiated in row-8 of EBR-II<sup>2</sup>. To augment these data, the same group is analyzing samples of  $^{233}\text{U}$  irradiated in row 4 of EBR-II. Results for about 40 nuclides should be available early in 1980. This group is currently preparing samples of  $^{233}\text{U}$  for irradiation in the Fast Flux Test Facility (FFTF) in late 1979. (See Section 4.)

3.3.3 Monoenergetic and High Energy Neutrons. Gindler and Glendenin are in the process of studying the systematics of fission yields verses neutron energy for  $^{233}\text{U}$  for the energy range of 0.1 to 8 MeV using a neutron generator. No results have been published to date. Fission yields for <sup>about</sup> 15 radionuclides (half-life in terms of days) for  $^{233}\text{U}$  irradiated with fission spectrum and 14.7 MeV neutrons are being measured in France by Laurec and Adams<sup>1</sup>.

#### 3.4 Uranium - 235

3.4.1 Thermal Neutrons. In late 1978, Maech, et al reported new mass spectrometrically <sup>electronically</sup> measured absolute fission yields for  $^{235}\text{U}$  thermal fission<sup>6</sup>. A program to measure relative yields based on mass spectrometric measurements is currently being conducted by Chitambar<sup>5</sup> and co-workers at Bombay, and is expected to be completed early 1980. In 1978, Strittmatter and Wehring<sup>21</sup> presented  $^{235}\text{U}$  thermal yields based on measurements using the fission-fragment recoil mass spectrometer, HIAWATHA. Several investigators<sup>25,29,30,31</sup> have or are in the process of measuring independent, fractional cumulative, and cumulative yields for  $^{235}\text{U}$  thermal fission. Isomer yield ratios are being measured by Reeder<sup>1</sup> using an on-line mass spectrometer. McKeon and Crouch are currently measuring the yield of tritium for  $^{235}\text{U}$  thermal fission.

3.4.2 Fast Reactor Neutrons. Absolute fast reactor fission yields for samples of  $^{235}\text{U}$  irradiated in row-8 of EBR-II were reported by Maech<sup>K</sup> and co-workers<sup>2</sup> in 1975. To augment these data, yields are also being measured for

samples irradiated in row 4 of EBR-II and should be available <sup>in</sup> early 1980. In both cases the measurement technique was isotope dilution mass spectrometry. These new data should aid in the continuing study<sup>8</sup> of variations in fast reactor fission yields with neutron energy. Davies and Sinclair<sup>22</sup> are currently in the process of irradiating samples in Prototype Fast Reactor (PFR) at Dounreay. The major emphasis will be absolute yields for the isotopes of neodymium. The measurement technique will be isotope dilution mass spectrometry. Radiochemical yield measurements for samples of <sup>235</sup>U irradiated in the core and blanket of the ZEBRA facility in the UK are being conducted by Cunninghame<sup>23</sup>. Also in the UK, Crouch is measuring fast tritium yields for samples irradiated in the same facility. Blachot and co-workers<sup>35</sup> have reported certain selected fractional independent yields for <sup>235</sup>U fast fission. Von Gunern is also engaged in measuring independent and cumulative yields for <sup>235</sup>U fast fission<sup>46</sup>. An irradiation in FFTF is scheduled by Mac<sup>K</sup> and co-workers to provide samples for establishing benchmark fission yields in FFTF. (See Section 4.)

3.4.3 Monoenergetic and High Energy Neutrons. Using a fast neutron generator, Glendenin and Staff are in the process of measuring radiochemical yields for <sup>235</sup>U over the energy range 0.1 to 8 MeV, as part of a program to better define the yield versus neutron energy correlation<sup>44</sup>. Fractional independent yields for several isotopes have been reported by Blachot<sup>35</sup> for <sup>235</sup>U fission with 3 MeV neutrons. Laurec and Adam<sup>1</sup> are measuring radiochemical yields of selected nuclides (half-life, days) on samples of <sup>235</sup>U irradiated with fission spectrum and 14.7 MeV neutrons.

3.5 Uranium - 236

The yield measurements for <sup>236</sup>U are rather limited. McKean and Crouch in the UK<sup>24</sup> are measuring tritium yields, <sup>and</sup> are Endal and co-workers<sup>42</sup> <sup>in the US,</sup> have reported yields for some short-lived gaseous fission products <sup>from</sup> degraded-fission-spectrum and 14 MeV neutron-induced fission.

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### 3.6 Uranium - 238

3.6.1 Fast Reactor Neutrons. Fast reactor yields for  $^{238}\text{U}$  have been reported by Maechl<sup>k</sup> and co-workers<sup>2</sup>. The measurement technique were isotope dilution mass spectrometry. Davies and Sinclair<sup>22</sup> are currently conducting an experiment in the PFR at Dounraey to provide samples for the measurement of  $^{238}\text{U}$  fast reactor fission yields. In this work, the emphasis will be yields for the isotopes of neodymium and the measurement technique will be mass spectrometry. Cunningham<sup>23</sup> is measuring radiochemical yields for a large number of nuclides for  $^{238}\text{U}$  irradiated in the core and blanket of the ZEBRA facility in the UK.

3.6.2 Monoenergetic and High Energy Neutrons. Debertjine has reported yields for  $^{238}\text{U}$  irradiated with neutrons from a  $^{252}\text{Cf}$  source<sup>34</sup>. Using a fast neutron generator the AIL group has reported  $^{238}\text{U}$  yields for the energy range 1 to 8.0 MeV<sup>10,11</sup>. Laurec and Adam<sup>1</sup> are measuring radiochemical yields on samples of  $^{238}\text{U}$  irradiated with fission spectrum and 14.7 MeV neutrons.

### 3.7 Neptunium - 237

Recently, Maechl<sup>k</sup> and co-workers<sup>5</sup> reported absolute fission yield data for  $^{237}\text{Np}$  irradiated in row-8 of EBR-II. The measurement technique was isotope dilution mass spectrometry, and the number of fissions was established by summing the number of atoms in the heavy mass peak. Blachot and Hospice have measured cumulative and chain yields for  $^{237}\text{Np}$  irradiated with a beam of filtered neutrons from the Melusine reactor. High resolution gamma ray measurements were made and the yields calculated relative to  $^{140}\text{Ba}$ . The results were contributed to the review paper on yields<sup>47</sup> at the meeting on Fission Product Nuclear Data, Petten, Netherlands, 1977.

### 3.8 Plutonium - 238

No significant <sup>yield</sup> data have been reported for  $^{238}\text{Pu}$  fission.



### 3.9 Plutonium - 239

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3.9.1 Thermal Neutrons. New  $^{239}\text{Pu}$  absolute thermal fission yields have been reported by Maeck and co-workers<sup>7</sup>. The measurement technique was isotope dilution mass spectrometry. Of particular importance is that the yields for the xenon isotopes and  $^{138}\text{Ba}$  are significantly different from previous yield data<sup>48</sup> reported by the same laboratory. These new data resolve several <sup>existing</sup> ~~previously-existing~~ problems in the evaluation of  $^{239}\text{Pu}$  thermal fission yields<sup>49</sup>. Chitambar and co-workers<sup>1</sup> at the Bhabha Atomic Research Center, are currently measuring relative  $^{239}\text{Pu}$  thermal yields using mass spectrometric techniques. Another group in the same laboratory<sup>37</sup> <sup>is</sup> are measuring radiochemical yields using mass spectrometric yields for using short lived nuclides using high resolution mass spectrometry as the measurement technique, and track detectors to determine the number of fissions. Dickens and McConnell, at ORNL, have recently measured radiochemical yields for  $^{239}\text{Pu}$  thermal fission. Reprints of the paper are available from the authors. Wehring and co-workers at the University of Illinois are measuring  $^{239}\text{Pu}$  thermal yields using a TRIGA reactor and the fission fragment mass spectrometer, HIAWATHA. Kaiser and Von Guentel<sup>26</sup> have reported cumulative mass yields for  $^{239}\text{Pu}$  irradiated with neutrons at the 0.3 eV resonance.

Independent and fractional cumulative yields for  $^{239}\text{Pu}$  thermal fission are being measured in several laboratories<sup>25,26,28,29,42</sup>. McKean and Crouch<sup>24</sup> are measuring the yield of tritium for  $^{239}\text{Pu}$  thermal fission.

3.9.2 Fast Reactor Neutrons. Absolute fast reactor fission yields for  $^{239}\text{Pu}$  irradiated in row-8 of EBR-II have been reported<sup>3</sup>. To augment these data, and to obtain more information relative to the change in yields with neutron energy, absolute yields for samples of  $^{239}\text{Pu}$  irradiated in row-2 of EBR-II are being determined in the same laboratory. The measurement technique is isotope dilution mass spectrometry. A new irradiation of  $^{239}\text{Pu}$  in FFTF is being planned to establish bench mark yields in FFTF. Davies and Sinclair<sup>22</sup> are currently irradiating samples

of  $^{239}\text{Pu}$  in PFR. Yields for the neodymium isotopes will be measured using mass spectrometric techniques. Cunningham<sup>23</sup> in the UK is irradiating samples of  $^{239}\text{Pu}$  in the core and blanket of the ZEBRA facility for the measurement of selected nuclides using radiochemical techniques.

3.9.3 Monoenergetic and High Energy Neutrons. Glendenin and Gindler<sup>44</sup>

at ANL are planning to measure the yields and change in yields of  $^{239}\text{Pu}$  irradiated with monoenergetic neutrons over the range of 0.1 to 8 MeV. Erdal and co-workers at LASL<sup>42</sup> have recently reported yields of selected short-lived isotopes of krypton and xenon for  $^{239}\text{Pu}$  irradiated with degraded-fission-spectrum and 14-MeV neutrons. Laruec and Adam<sup>1</sup> in France are completing the radiochemical measurements for determining the cumulative yields of several radioisotopes with half-lives in the order of days. The number of fissions is being established by use of a fission chamber.

3.10 Plutonium - 240

3.10.1 Fast Reactor Neutrons. Recently Maechi and co-workers<sup>4</sup> reported

absolute fast reactor fission yields for  $^{240}\text{Pu}$ . The measurement technique was isotope dilution mass spectrometry and the number of fissions are based on the sum of the atoms in the heavy mass peak. Davies and Sinclair are irradiating samples of  $^{240}\text{Pu}$  in PFR. Absolute yields, using mass spectrometric analysis, are to be determined for the isotopes of neodymium,  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$ , and  $^{144}\text{Ce}$ . McKean and Crouch<sup>24</sup> are in the process of measuring the fast fission yields of tritium from samples of  $^{240}\text{Pu}$  irradiated in the ZEBRA facility.

3.10.2 Monoenergetic and High Energy Neutrons. Methaway and co-workers<sup>13,14</sup>

have reported  $^{240}\text{Pu}$  yields for samples irradiated in a degraded-fission-spectrum and 14.8 MeV neutrons. The measurement technique was high resolution gamma ray spectrometry.

3.11 Plutonium - 241

3.11.1 Thermal Neutrons. Relative yields for about 20 mass numbers are being determined employing mass spectrometric techniques by Chitambar and co-workers at the Bhabha Atomic Research Center<sup>1</sup>. Recently Dickens<sup>2, 15</sup> reported <sup>241</sup>Pu thermal yields for 17 short-lived fission products based on high resolution gamma spectrometry measurements of microgram samples of <sup>241</sup>Pu irradiated for ~/min. *following the thermal neutron irradiation*

3.11.2 Fast Reactor Neutrons. Absolute fast yields for <sup>241</sup>Pu irradiated in row-8 of EBR-II have been reported by Maeck and co-workers<sup>3</sup>. The measurement technique was isotope dilution mass spectrometry. Davies and Sinclair<sup>22</sup> are irradiating samples of <sup>241</sup>Pu in PFR for the measurement of the yields for the isotopes of neodymium, <sup>90</sup>Sr, <sup>137</sup>Cs, and <sup>144</sup>Ce, using mass spectrometric techniques. *The Idaho group has prepared* Samples of <sup>241</sup>Pu have been prepared for irradiation in FFTF to establish <sup>241</sup>Pu bench mark fission yields in FFTF *(see Section 4)*. *Crane* McKean and Gronich are planning to measure the yield of tritium for <sup>241</sup>Pu fast fission.

3.12 Plutonium - 242

3.12.1 Fast Reactor Fission Neutrons. *In 1977,* Recently Maeck and co-workers<sup>4</sup> have reported fast reactor yields for <sup>242</sup>Pu irradiated in row-8 of EBR-II. The measurement technique was isotope dilution mass spectrometry.

3.13 Americium - 241, 243

Recently, experiments were concluded at the Lawrence Livermore Laboratory relative to the measurement of the fission yields of <sup>241</sup>Am irradiated with slightly degraded fission spectrum and 14.8 MeV neutrons<sup>50</sup>. Radiochemical measurements were performed for about 50 fission products. The results are currently being prepared for publication.

by the Irbako group.

Samples of  $^{241}\text{Am}$  and  $^{243}\text{Am}$  <sup>were</sup> have been irradiated in row-8 of EBR-II A

Initial studies on the  $^{241}\text{Am}$  show large amounts of stable fission product

contamination in the target material. At this time it is highly questionable

that <sup>in all, not</sup> reliable yield values will be obtained from these samples.

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#### 4. FISSION YIELD MEASUREMENT PROGRAM FOR FFTF

A program has been started in the U.S. to measure fast reactor fission yields <sup>from</sup> for samples of several fissionable nuclides irradiated in the Fast Flux Test Facility (FFTF), at Hanford. This program, which is an extension of the previous experiments<sup>2,3,4,5</sup> conducted by the fission yield measurement group at Idaho, is multipurpose with the following objectives: 1) to provide a group of heavy element reference standards for which the number of fissions and burnup is well known, and to which other experiments conducted in the same irradiation can be compared, 2) to provide fission <sup>ratio</sup> data for several different fissionable nuclides in FFTF for various locations within the core, 3) to verify fast yields measured in EBR-II, and 4) to provide benchmark fission yield data for FFTF. The samples will be irradiated during the 8-day high power core characterization experiment for FFTF. The irradiation will be highly instrumented and many dosimetry packets supplied by the Hanford Engineering Development Laboratory (HEDL) will be incorporated in the experiment to provide detailed spectral information.

~~Samples of highly enriched oxides of  $^{233}\text{U}$ ,  $^{235}\text{U}$ ,  $^{239}\text{Pu}$ , and  $^{241}\text{Pu}$  have been blended with spectral information.~~

Samples of highly enriched oxides of  $^{233}\text{U}$ ,  $^{235}\text{U}$ ,  $^{239}\text{Pu}$ , and  $^{241}\text{Pu}$  have been blended with high purity nickel powder and encapsulated in nickel capsules by the fission yield measurement group at Idaho. After irradiation, which should occur in early 1980, the samples will be allowed to "cool" several months and then returned to Idaho for analysis.

Duplicate samples of each fissionable material will be placed at or near reactor mid-plane in rows 1, 2, 4, 5, and 6 in the FFTF core. Row-6 is the outer edge of the core. The samples in rows 1 and 6 contain approximately one gram or more of fissionable material and those in rows 2, 4, and 5 about 50 mg of target material. Absolute fission yields for the bulk of the fission products

will be measured on the samples from rows 1 and 6 to establish benchmark yield data, and to determine the change in the yields as a function of neutron energy. The number of fissions will be based on the summation technique. The measurement technique will be isotope dilution mass spectrometry. It is expected that <sup>each sample will produce</sup> about  $10^{19}$  fissions ~~will occur in these samples~~ and that the yields <sup>should be within</sup> ~~can be established with~~ an uncertainty of about 1%, relative. The smaller samples in rows 2, 4, and 5 will be analyzed for the isotopes of neodymium, and  $^{137}\text{Cs}$ , and possibly a few other isotopes. Using the fission yield data generated from the row-1 and -6 samples, fission rates will be established for the other samples.

Also to be evaluated, will be the ratio of  $^{150}\text{Nd}/^{143}\text{Nd}$ . Previous experiments<sup>2</sup> have shown this ratio be be highly correlated with the spectral index,  $^{238}\text{U} (n,f)/^{235}\text{U} (n,f)$  and hence, may be useful as a new spectral index.

Currently, it is expected that the irradiation will be performed early in 1980 and that the analytical work will start in late 1980. The primary emphasis will be on the plutonium samples.

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