Fission Product Yields as a Diagnostics for Plutonium Burnup

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Abstract

I describe progress made in recent years in determining fission product yields for fast neutron reactions on plutonium. Discrepancies of the order of 5–10% have been partially resolved, allowing fission burnup to be determined to a few percent accuracy.

1 Introduction

This paper represents an extract from “Fission Yields and Other Diagnostics for Nuclear Performance”, LA-UR-12-00727 (2012): a talk on the occasion of receiving the E.O. Lawrence Award, and focuses on one aspect of that work - fission product yields.

Until recently Los Alamos and Livermore disagreed on the yields in kilotons assigned to a plutonium nuclear explosion. This wasn’t always the case. Since the earliest days of nuclear science, Los Alamos developed methods to determine the yields from measurements of the fission products (FP) in the debris after the explosion, through use of calibrated laboratory experiments involving fission chambers inside critical assemblies. These critical assemblies allowed radiochemists to determine exactly how to translate the beta decay radioactivity of the fission products to the number of fissions that occurred, and hence to the yield. Originally Livermore followed Los Alamos’ approach, and the labs were on a consistent “fission basis”. Later, in the 1970s, Los Alamos repeated the calibration experiment using a critical assembly that better mimicked our applications (creating a fast spectrum, not a thermal neutron spectrum) as part of the Inter-Laboratory Reaction Rate (ILRR) collaboration, and found a different result compared to its original 1950s result: the key $Q_{99}$ value \[ Q_{99} \] for $^{239}\text{Pu}$ was determined to be 1.015 instead of 0.966. The reason for the discrepancy was thought to be due to a self-shielding problem in the early experiment, and adopting the new experimental results led Los Alamos to lower its fission yield assessments. However, Livermore had – we concluded – remained on the original basis, so that for the last two decades the Laboratories have had an offset in their assessments. Livermore felt it prudent to be cautious before making a change to again be consistent with LANL. In part this was because, for the key fission product we use as an indicator (neodymium-147, $^{147}\text{Nd}$) the fission product yield in use at Livermore happened to be in good agreement with an independent fast reactor measurement from Idaho National Laboratory by Maeck [2] (as I’ll explain later, this puzzle was resolved by the identification of a neutron energy-dependence to the $^{147}\text{Nd}$ product yield).

Thus, we had a situation in the 1990s and 2000s where the equivalent sets of specialists at LANL and LLNL knew their results were different, but each group felt their results were correct! Additionally, Livermore had moved to use a different approach for much of their fission product work that involved measuring fission products using Germanium detectors for the decay gamma-rays, together with use of fission product yields. These methods differed from Los Alamos’ more traditional beta decay radiochemical methods, and for a while at least some of the differences between the labs was due to different languages used between the specialists. LANL continued to use the somewhat archaic radchem language of “K factors and R and Q values” [3], that had its origins in the Manhattan project, while Livermore migrated to the more widely used “fission product yield”. Only through numerous exchanges between the labs, and Don Barr’s writings was it understood that the approaches are equivalent when care is taken in determining the physical constants.

The confusion continued. The Livermore radiochemists from the nuclear testing era had mostly retired and few records remained documenting the origins of their FPYs that were being used in their
yield assessment work. Livermore pointed to examples of their FPY evaluations based on measurements published in the open literature. But it was shown that, for the key fission products used in yield assessments such as $^{147}$Nd, $^{95}$Zr, $^{99}$Mo, $^{144}$Ce, etc., the values Livermore used almost certainly came from the same traditional methodology ("FPY= FPY-235-th. $Q_{99}$. R") used at LANL. This is no surprise since it reflects a desire for continuity in yield assessments made over the years. But it also showed why Livermore FPY values in use until recently were offset compared to LANL’s: they used the old (deficient) 1950s LANL $Q_{99}=0.966$ value.

The challenge I faced was to assemble evidence that would elucidate what are the correct FPY values for fast neutrons on plutonium, and explain why LANL and LLNL values differed. The previous paragraphs summarize our conclusions on why the labs differed, but work was needed to determine which values were most correct. It was not enough to say that the esteemed radiochemists from Los Alamos – Barr, Knobeloch, and so on – undertook the LANL-ILRR 1970s experiments and obviously measured the key quantities correctly, or that the successive generations of excellent LANL researchers – Mac Innes, Inkret, Wilkerson, Selby, Keksis, Burns, Meade, Wallstrom etc – had carefully analyzed these data and found them to be trustworthy! Livermore could validly ask why we should trust this particular experiment, which had not (at that time) been documented in the open literature? And although we showed [4] that results from the 1970s LANL-ILRR experiment were in excellent agreement with many other accurate and independent measurements published in the literature (e.g. Maeck [2]; ILRR [5]), this was not the case for the key $^{147}$Nd FPY. Here, Maeck’s measurement agreed instead with Livermore’s value (like LANL’s historic 1950s value) and not the LANL-ILRR value that LANL is now recommending! Why was this?

There were two particular advances I made that helped solve this problem, described below: (a) an identification of a subtle energy dependency of the key $^{147}$Nd FPY; and (b) use of a meta-analysis to expand our knowledge base of information on the magnitude of FPYs; the result of this analysis supported the validity of LANL’s measured values.

### 2 Energy dependence of FPYs

I came to the conclusion that the apparent discrepancy between the LANL FPY data for $^{147}$Nd and Maeck’s fast-reactor $^{147}$Nd value is due to the different neutron spectra in the two experiments: fast reactors have an average neutron energy of a few hundred keV, whereas the LANL data (appropriate for our applications) have an average energy closer to 1.5-2 MeV, and there are physical reasons why the $^{147}$Nd FPY can have a positive energy dependence over this region. Thus, both the LANL and the Maeck values can be correct within their uncertainties; they just apply to different energy regimes.

It is well known that FPYs often have neutron energy dependencies. The mass distribution of FPYs is double-humped, owing to shell effects which favor a heavy peak near the closed-shell A=132 (plus the few extra nucleons captured back after the rupture of the neck between the two fragments, giving a peak at about A=135), and consequently a light peak near 102 (this comes from 240, the initial compound system mass before fission, – 135 for the heavy fragment – 3, the average number of prompt fission neutrons emitted). FPYs in the valley (near A=120) increase with incident energy as the symmetric fission breakup mode becomes energetically more possible, and the FPs in the wings also increase with energy, whilst the yields for the FPs at the peaks decrease slightly with energy. The FP indicators used by LANL and LLNL were chosen to be roughly energy-independent, lying nearer the peaks of the double-humped FPY distribution. And the lore at the labs had been that these FPs are energy independent. Whilst this is approximately true, at the few percent level energy dependencies can occur for all FPs, including $^{95}$Zr, $^{144}$Ce, and $^{147}$Nd.

Fortunately LANL measured a rich database of 17 plutonium $^{147}$Nd R-values within critical assemblies with varying neutron spectra, all the way from thermal up to fast energies (the hottest assembly with the highest energy neutrons being Jezebel, a sphere of plutonium). This allowed me to develop an
independent check of the $^{147}$Nd FPY energy dependence since the FPY is proportional to the R-values, and indeed we do see a positive variation between the softest and hottest assemblies, supporting the hypothesis. The effect is subtle, though, since the energy-dependence magnitude (3–4 % relative change over an MeV) is approximately the same as the variance of the data. Trends in the systematic behavior of FPY energy dependencies for the whole mass range of FPs, based on numerous independent experiments, build a phenomenological picture of the energy dependencies that makes physical sense, and $A=147$ is seen to be at the transition between negative and positive dependencies for plutonium (just on the positive side – see Fig.10 of Ref. [4]). My colleague John Lestone has recently developed a theoretical underpinning of these dependencies based on a model for fission [6], and his results support these conclusions.

Subsequent independent Livermore studies of the experimental $^{147}$Nd FPY data (Thompson et al., Ref. [7]) have led to a similar energy-dependence result ($\sim 3.2\%$ per MeV). Dardenne of Livermore also developed another way of viewing the data based on taking FPY ratios to other FPs (so that certain systematical uncertainties cancel). This approach also confirms the $^{147}$Nd FPY positive energy dependence over the 0.5-2 MeV range – when observing the $^{147}$Nd FPY in ratio to $^{140}$Ba (which we think is essentially energy-independent over this range) Thompson and Dardenne found an energy-dependence of about 4.5%-per-MeV [7], consistent with our result. A separate group of consultants commissioned by Livermore, led by Stan Prussin [8], studied the problem from a different and rather clever perspective (see also Maeck [9]), focusing only on the isotope dilution mass-spectrometry reactor data and again found a similar result (2.4–4.0%-per-MeV) for the $A=147$ energy-dependence.

FPY energy dependencies would be of little practical concern if significant uncertainties in our plutonium yield assessments could be tolerated, as was the case for much of the nuclear testing period. But now that our accuracy goals are much higher – of the order of a few % – such phenomena need to be considered.

3 Magnitude of fission product yields determined through a meta-analysis

Although the energy dependence hypothesis was able to explain part of the difference between LANL and LLNL’s yield values, an important question remained to be settled: how sure can we be that LANL’s overall FPY magnitudes are correct? Until the recent consensus between the labs there was a general offset in magnitude for all the important FPYs we use – $^{95}$Zr, $^{99}$Mo, $^{140}$Ba, $^{144}$Ce, as well as for $^{147}$Nd. Energy dependence considerations weren’t particularly relevant to resolving this discrepancy.

$^{99}$Mo is LANL’s standard fission product – all other FPs are measured in ratio to $^{99}$Mo (thus, $^{99}$Mo is analogous to carbon in the field of atomic masses). The absolute scale of all of LANL’s fast neutron+Pu FPY values, including $^{147}$Nd, is therefore set by the $^{99}$Mo $Q_{99}=1.015$ measurement in the seminal LANL-ILRR experiment. Livermore’s values differed by an offset compared to this value, arising – we believed – from their use of the historic $Q_{99}=0.966$. I therefore sought to find independent information that would elucidate the correct value for $Q_{99}$ to resolve this difference. But this was complicated by the fact that for $^{99}$Mo there is a dearth of experimental information beyond LANL’s measurements. One of the few other sets of reported $^{99}$Mo fast neutron on plutonium FPY data came from Maeck [9], and agreed with LANL, but was interpolated from nearby nuclides. The other direct measurement, by Laurec [10], was discrepant with LANL’s data – though all of Laurec’s FPY data for fission-spectrum neutrons on plutonium tend to lie below other laboratory’s measurements, a discrepancy we still do not understand.

I realized that the large suite of Los Alamos R-value measurements contained a hidden treasure of information on $^{99}$Mo, it just needed to be teased out. The R-value measured data for various fission products $j$ contains ratio information on the production of the fission product $j$ to that of $^{99}$Mo. (Actually it is a ratio of ratios, see Refs. [3, 4]). These other fission product $j$ FPYs are often known accurately from a variety of independent measurements published in the literature. Thus by using these other FPYs together with LANL’s R-value ratio data, the $^{99}$Mo FPY can be inferred. By following this prescription...
I determined $^{99}$Mo values, and found a consistent *meta-analysis* result $Q_{99}=1.019\pm0.8\%$ that supported LANL’s direct measurement of $Q_{99}=1.015\pm2\%$ [4].

A meta-analysis is defined as procedure by which multiple data sets set can be combined to better determine a quantity, to overcome problems of small sample sizes. This well describes the above process, where information embodied in R-values was used to expand our knowledge of the $^{99}$Mo fission product data. By expanding the available database from 1 to N values, the uncertainty on the FPY result was reduced by $1/\sqrt{N}$. The result I obtained for $Q_{99}$ further supported LANL’s yield assessments, and reduced the uncertainty.

Finally it is worth noting that in the two years that have passed since I published the meta-analysis results, a consistency check can be done based on new evaluated $^{99}$Mo FPY data that have been published by LANL, LLNL, and by Prussin (see Table 1). This is done by computing $Q_{99}$ from them by dividing the plutonium fission-spectrum FPY by the ENDF FPY for thermal reactions on uranium-235. When one does this, very good agreement is seen (LANL, 6.23/6.108=1.020; Prussin, 6.22/6.108=1.018; LLNL, 6.19/6.108=1.013; the average of these 3 is 1.017 which compares well with $Q_{99}=1.019\pm0.8\%$ from my meta-analysis).

Livermore has now conducted an independent evaluation effort on the magnitude of the FPY for fissile spectrum neutrons on plutonium (Thompson *et al.* [7]). Unlike the older Livermore values, which were typically 5–9% lower than LANL’s values, the magnitude of their new results agree with ours (within about 2% or better), see Table 1.

4 Conclusions

In this paper I haven’t described the (less important for many applications) 14 MeV range, where the laboratories still have some significant differences. At Los Alamos we have refined our understanding of our 14 MeV experiments and evaluations, and we recently documented our results in peer-reviewed open publications [12, 13]. But the resolution of our present differences at 14 MeV will require future experiments; see below.

There are some remaining puzzles to be solved, including unknown reasons for the systematically lower plutonium FPY measured by our excellent CEA colleagues [10]. We have initiated various experiments that aim to corroborate (or who knows, maybe invalidate?) our present understanding: new measurements being planned in Nevada using our critical assemblies will look at FPY energy dependencies between thermal and ~1.5 MeV (thanks to Bob Little, Todd Bredeweg, and others in the Criticality Safety community); a detector being fabricated at LANSCE under an LDRD/DR project (Morgan White, Frederick Tovesson) will map out trends of FPY for all masses and energies; and measurements at TUNL through the NNSA Stewardship Science Academic Alliance program (with Jerry Wilhelm, John Becker, Anton Tonchev, David Vieira, Mac Fowler, Mark Stoyer *et al.*) are looking at specific FPY ratios from fast to 14 MeV energies. What started as an effort to resolve a longstanding discrepancy between LANL and LLNL has evolved into the re-vitalization of fission physics research.

References

[1] $Q_{99}$, for $^{99}$Mo, is the ratio of the fission product yield for fission spectrum neutrons on plutonium to that for thermal neutrons on $^{235}$U. At Los Alamos it was not determined experimentally from ratios of FPY per se, but rather from ratios of count rates of the FP radioactive decay.


Table 1: Summary of FPYs from fission spectrum neutrons on $^{239}$Pu in units of % per fission. The values are quoted for an average energy causing fission of 1.5 MeV (except for the $^{99}$Mo data that are quoted at the relevant energy of the Bigten molybdenum experiment, $\sim$0.5 MeV).

<table>
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<th></th>
<th>LANL-ILRR exp.</th>
<th>LANL-eval</th>
<th>Prussin eval.</th>
<th>LLNL-eval. (new)</th>
<th>LLNL-eval. (old)</th>
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<td>$^{95}$Zr</td>
<td>4.80±0.18</td>
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<td>$^{99}$Mo</td>
<td>6.20±0.15</td>
<td>6.23±0.04</td>
<td>6.22±0.12</td>
<td>6.19±0.11</td>
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<tr>
<td>$^{144}$Ce</td>
<td>3.68±0.13</td>
<td>3.62±0.08</td>
<td>3.66±0.05, 3.67±0.05</td>
<td>3.69±0.02</td>
<td>3.34</td>
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<tr>
<td>$^{147}$Nd</td>
<td>2.09±0.05</td>
<td>2.10±0.03</td>
<td>2.13±0.04, 2.11±0.04</td>
<td>2.05±0.06</td>
<td>1.97</td>
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