RAPID AUTOMATED NUCLEAR CHEMISTRY FACILITY FOR THE STUDY OF SHORT LIVED FISSION PRODUCTS

> R. A. Meyer, E. A. Henry, O. G. Lien, H. G. Hicks and P. C. Stevenson\* Lawrence Livermore Laboratory

We have known fission for over four decades, yet it continues to present challenging problems both as to its fundamental nature as well as to its optimal use as an energy source. Many of these problems can be attacked if the fission products are studied in detail. For years the long-lived fission products, which can be isolated by traditional chemical techniques, have been investigated. However, we were quite ignorant of the short-lived nuclides with halflives up to a few hundred seconds, mainly because the techniques of chemical isolation have required times far in excess of the decay half-lives; hence, these nuclides decayed before they could be measured.

To study short-lived fission product nuclei in detail, we have developed techniques of rapid chemical isolation and spectroscopy for these fission products. Because of the very short half-lives, the useful measurement period resulting from a single chemical isolation is very short, and only a small fraction of the necessary data can be obtained by an individual measurement. We therefore repeat exactly the same measurement many times to build the statistical base. Furthermore, since the nuclide species of interest represent a small fraction of the total radioactivity, we separate that nuclide from the highly radioactive collection of fission products by remote means. Our facility includes both an automatic batch system (autobatch) and a system for continuous isolation of single-element fission products.

We have developed, in particular, rapid techniques to separate fission-product isotopes of the group V-A and VI-A elements. We use gaseous hydride separation and transfer techniques for the antimony and arsenic elements. For bromine we have isolated its precursor, selenium, followed by extraction of the daughter bromine, allowing enhancement of 55.5-s  $^{86}$ Br over 56.1-s  $^{87}$ Br by more than a factor of 25, which compares with the approximate ratio of 1 produced by isolation of bromine from fission products. The isolation of tellurium from fission products has been done by a combination of hydride production followed by extraction in xylene. The range of half-lives we study for the Sb isotopes range from 11-s to a few minutes. The complexity of studying the As and Br isotopes is made difficult by the similarity in half-lives of  ${}^{86}$ Br and  ${}^{87}$ Br as well as 16.5-s  ${}^{80}$ As, 19-s  ${}^{82}$ As<sup>m</sup>, 13.3-s  ${}^{82}$ As<sup>g</sup>, and 13.5-s

<sup>83</sup>As.

## AUTOBATCH

We use a pneumatic tube to transport samples rapidly in and out of the Livermore Pool-Type Reactor (LPTR). Next, we extract the fission products from the transportation container (rabbit) and chemically isolate the element of interest. Once it is isolated, we must quickly measure its characteristic radiations before its level of activity drops too low.

We have had to improve considerably our original system, which was initially designed for the isolation and study of 2-min activities. Characteristics of the improvements are compared in Fig. 1 with the

Lawrence Livermore Laboratory

original system; the improvements have come from our effort to automate fully both the chemical separation and the spectroscopy measurements.

The full cycle of operation for our automated system is diagrammed in Fig. 2. Rabbits containing doubly encapsulated 1-mg samples of U-235 in solution are shot into the reactor from an automatic rabbit launcher. A computer controls the loading and firing from a magazine that holds 38 rabbits. A microprocessor loads and sends the rabbit through a pneumatic tube (activated by nitrogen gas) to the core of the LPTR. The sample is irradiated for a specific time, then the microprocessor sends it through a second pneumatic tube to a receiver located in a chemistry laboratory hood approximately 150 ft from the core of the LPTR. Once emplaced in the laboratory, the rabbit is punctured by a double concentric needle that extracts the fission product, adding acid and carrier (a trace amount of antimony) in the process.

Purging with  $N_2$  gas (see Fig. 2b) cleans out

the gases produced during fission and precludes their daughter activities from being observed in subsequent spectroscopy measurements. The actual chemical separation of Sb from the other fission products is made upon addition of sodium borohydride (NaBH,)

to the acidified fission products and carrier. The stibene and arsine (gaseous  $SbH_3$  and  $AsH_3$ ) is

transported through a  $CaSO_4$  trap that eliminates other

hydrides produced by NaBH4 raction. The final step is

destruction of the stibene with KOH in ethanol; the homologue AsH3 passes through. This final step leaves

the Sb on a substrate in front of a radiation detector. As soon as the final chemical step is finished, the microprocessor turns on the spectrometers for measurement.

An important feature of the microprocessor control is the capability for simultaneous operation; for example, spent reagents are cleaned from the chemistry system and new reagents replenished during the measurement period (Fig. 2). This reduces the cycle time and, hence, increases the number of measurements that can be made in an operational day. The actual number of measurements, then, depends only on the length of the measurement segment, which can vary from a short time - if gamma-gamma-time-threeparameter coincidence measurements are being made on a short-lived activity such as 10-2 Sb-134 decay - to 30 min in our experiments to measure the independentfission-yield isomer ratio of <sup>132</sup>Sb<sup>g</sup> vs <sup>132</sup>Sb<sup>m</sup>. The measurement rate has been greater than 1000 isolation/ measurement cycles in one 10-hour LPTR operating day.

We have developed a number of novel chemical techniques in our efforts to isolate rapidly some fission-product elements. For example, we have based some of our chemical isolations on the generation, transport, and destruction of the gaseous hydrides of class V-A and VI-A elements. We discovered that the mere injection of a caustic (NaOH) into the transport line isolated nearly pure As (see Fig. 3). (Although we had been able to obtain excellent antimony separation on the time scale of 0.5 to 1 min using this same hydride technique, we found that the kinetics of ion exchange were too slow to be useful for the isolation of 10-s <sup>134</sup>Sb.) Also, we found that

<sup>\*</sup>Authors' Affiliation and Address:

Livermore, California 94550

tellurium hydride could not be isolated by the gasgeneration technique when a mass of tellurium carrier (typically 1 mg per separation) was added. However, we did obtain a good tellurium separation after we discovered that, in the gas phase, a massive amount of tellurium will polymerize, much as its homologue sulfur does.

The study of some nuclides requires chemical separations on a time scale much less than their half-lives. For example, not until we were able to perform 10-s Sb separations were we able to study 850-s (12.5-min)  $^{133}\text{Te}^{\text{g}}$  by itself. Several tellurium isotopes have half-lives in the range of 10 to 40 min. In order to isolate only  $^{133}\text{Te}^{\text{g}}$ , we had first to isolate antimony, allow it to decay, and separate

to isolate antimony, allow it to decay, and separate the daughter tellurium. By selective timing of the separation steps, nearly pure  $^{133}$ Te<sup>g</sup> was obtained.

Since we must repeat our experiments for statistical accuracy, we want to assess the experiment's progress while we are taking data. Consequently, we have developed spectrometers that have the capability of on-line analysis of the data.

One such spectrometer<sup>2</sup> allows nearly instant analysis of gamma-gamma coincidence spectra, instead of having a 2- to 7-day delay.

### DATA APPLICATIONS

## Nuclear Power

In designing a power reactor, provisions must be made for several categories of hypothetical accidents. For example, if an accident causes the loss of the cooling water, automatic controls would instantly drop the reactor control rods back into the core, thereby stopping operation of the reactor and any further fission-product production. However, residual fission products holding a large amount of energy would remain. This energy would be converted into heat at a rate governed by the half-lives and in an amount governed by the decay energies of all the fission products present.

Our first measurements for this application have been on the properties of the short-lived antimony isotopes  $4.2-\min^{132} \text{Sb}^m$ ,  $2.4-\min^{132} \text{Sb}^g$ ,  $2.3-\min^{133} \text{Sb}$ , and 10-s <sup>134</sup> Sb. Our results show that even previously accepted average gamma-decay energy data are incorrect by as much as 45%. In Table 1, we compare our results with previous work.

Table 1. Changes in average γ-ray energies of isotopes associated with reactorsafety decay-heat isotopes.

	Isotope			
	132 <sub>Sb</sub> m	132 <sub>Sb</sub> g	<sup>133</sup> Sb	<sup>134</sup> Sb
Previous E <sub>Y</sub>	2.344	2.488	1.874	2.086
Present $E_{\gamma}$	2.799	2.535	2.531	2.632
% Change	19	1.9	45	26

The mass-133 fission product chain is particularly important in studying the fission process via measuring independent yields. Unfortunately, little has been known of the mass 133 chain exact properties. For example, literature values for the chain branching at antimony to tellurium vary by almost a factor of 100. In the past few years we have determined all the absolute values for the decay properties in the chain using our system. Our results are shown in Fig. 4. Our first experimental data<sup>4</sup> showed several unsuspected features in the properties of the decay chain that have been confirmed by recent measurements. Particularly important are:

• The absolute intensity of the gamma rays from the  $^{133}$ Sb decay.

- That 10% of all  $^{133}$  Te<sup>m</sup> decays go through a
- 9-s 19/2<sup>-</sup> isomer in <sup>133</sup>I at 1634 keV.
  That the 334-keV gamma-ray intensity originally thought to represent the isomeric transition for <sup>133</sup>Te<sup>m</sup> decay to <sup>133</sup>Te<sup>g</sup> only represented 60% of the intensity.<sup>4</sup> The remaining 334-keV gamma-ray intensity belongs to the unsuspected dominance of lower-energy transitions at high

excitation energy in 133I.

These features are important for using the gammaray data to determine the yields of the early mass-133 fission-product chain. They will be used as the basis for our fission-yield measurements.

We have adopted a large-basis shell-model code that has been successful in describing nuclei that range from the light closed-shell nuclei to those in the  $\underline{Z}$  = 50,  $\underline{N}$  = 82 region.<sup>5</sup> After testing gross nuclear properties, we have compared in detail the code's predictions against our results on the energy level properties of  $^{132}$  Te,  $^{133}$  Te,  $^{134}$  Te, and  $^{135}$  Xe. As an example, in Fig. 5, we compare our calculational and experimental results for <sup>134</sup>Te levels (as populated by the decay of 10-2  $^{134}$ Sb). From these results we have discovered an unsuspected influence on the level properties of this closed-shell nucleus. Calculations with only the valence nucleons beyond shell closure fail to replicate the transition rates of the nuclear level decays. Only by allowing the unsuspected influence of the closed-shell core nucleons were we able to account for the experimental results.<sup>5</sup> Our measured value of the  $6_2^+ \rightarrow 6_1^+$  versus  $6_2^+ \rightarrow 4_1^+$ 

transition was more than an order of magnitude different from the value predicted without the influence of the core nucleons.

#### References

<sup>1</sup>The Livermore Pool-Type Reactor is described in Lawrence Livermore Laboratory's <u>Evergy and Technology</u> <u>Review</u> for June 1977 (UCRL-52000-77-6), p.1.

- <sup>2</sup>W. D. Ruhter, D. C. Camp, L. G. Mann, J. B. Niday, and P. D. Siemens, <u>A Megachannel-Coincidence System</u> <u>Using a PDP-8/E Computer and Moving-Head Disks</u>, Lawrence Livermore Laboratory, Rept. UCRL-78221 (1976); and W. D. Ruhter, Ph.D. thesis, University of California, Davis (1978).
- <sup>3</sup>G. Skanemark, Ph.D. thesis, Chalmers University, Göteborg, Sweden (1977).

<sup>4</sup>R. A. Meyer, <u>Problems in Vibrational Nuclei</u>, G. Alaga, V. Paar, and L. Sips, Eds. (North Holland Pub. Co., Amsterdam, 1977), p. 128.

<sup>5</sup>S. M. Lane, Ph.D. thesis, University of California, Davis (1978).

# Proceedings of the Eighth International Conference on Cyclotrons and their Applications, Bloomington, Indiana, USA



Fig. 1. Improvements in autobatch system.





Fig. 2. Timing diagram for autobatch system 2a (upper figure) overall sequence; 2b (lower figure) chemistry sequence for As chemistry.







Fig. 4. Decay properties of mass 133 Fission product chain. Numbers next to slanted arrows represent % decay of that isotope (e.g. 133Sb decays 82% to 133Teg and 18% to 133Tem). Numbers above vertical arrows give the major γ-ray energy and % intensity (e.g. the 1096 keV γ-ray represents 42% of all 133Sb decays). Lower left insert shows change from previous values to our current values for 133Sb decay while upper insert shows changes for 133Tem decay.



Fig. 5. Comparison between our experimental results and large basis shell model calculation. Lower bargraph insert compares the observed and calculated branching ratio for the  $6_2^+ + 4_1^+$  to  $6_2^+ + 6_1^+$  transitions. Calc. I is without core nucleons. Calc. II is with core nucleons.