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NUCLEAR DATA MEASUREMENTS OF NEUTRON-RICH NUCLIDES PRODUCED IN FISSION USING ON-LINE ISOTOPE SEPARATION

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Abstract: The use of the technique of mass separation coupled on-line to a source of fission-products has in recent years provided new information on the nuclear decay properties of such nuclides. In addition to their relevance to nuclear physics, these data also have important applications in both fission reactor technology and nuclear astrophysics. In this paper, we discuss nuclear measurement activities at the INEL pertinent to these latter two fields using ISOL facilities at the INEL and at BNL.

(radioactivity, half-lives, Q values, beta decay, mass separation)

Introduction

The fission process produces a wide range of neutron-rich nuclides far off the line of beta stability. Use of mass separation coupled on-line to a fission source has provided a new method for systematic study of the nuclear decay properties of these fission isotopes.

In addition to their importance in studies of basic nuclear properties of neutron-rich nuclei, fission products, because of their intimate link with energy production in fission reactors, occupy a unique position in the field of applied nuclear decay data. As discussed in Refs. 1 and 2, interest in fission product decay data has increased markedly in recent years. In addition to their critical role in nuclear reactor technology (decay heat source term, environmental hazards, etc.), such data are necessary to allow refinement of nuclear models of very neutron-rich nuclides used in astrophysical calculations involving the rapid neutron-capture process (r-process) of elemental synthesis in stellar environments[3].

In this paper, we discuss recent nuclear data measurement activities at the INEL pertinent to both fission reactor technology and to nuclear astrophysics. For these studies, we are using ISOL facilities both at the INEL and BNL (TRISTAN). At the INEL ISOL facility we are presently undertaking a systematic study of the gross nuclear decay properties of short-lived fission-product isotopes (i.e., ground-state half-lives, beta-decay energies and beta-strength functions). At TRISTAN we are undertaking measurements of delayed-neutron energy spectra of those fission-product precursors most important to reactor technology.

The INEL ISOL Facility

The ISOL facility at the INEL is unique in its use of spontaneous fission of ^{252}Cf as the source of fission isotopes. In designing this facility, it was anticipated that large electro-deposits (up to ~1 mg) of ^{252}Cf would be needed to allow for high-quality nuclear spectroscopy. Thus, it was necessary to locate the ^{252}Cf fission sources in a hot cell and this in turn has mandated the use of a gas-jet transport arrangement for coupling the fission products on-line to the mass separator. A schematic of the ^{252}Cf-based ISOL facility is shown in Fig. 1. In this facility, which is described in Ref. 4, the ^{252}Cf sources (electrodeposited at ORNL) are mounted in a pressuried chamber which forms an integral part of the gas-jet transport arrangement. After thermalization, and attachment to the NaCl aerosols seeded in the He-gas flowing through the chamber, the fission fragments are transported with high efficiency (>50%) into the ion source. Ion sources used to date have been of the electron bombardment heated hollow cathode design. Following mass separation, the selected mass fraction is collected on a tape which can be moved in either of two transport lines to an appropriate detector station.

Ground-State Half Lives

Using the INEL ISOL facility, we recently undertook a systematic search in the mass region A>150 to observe new rare-earth fission-product isotopes. A result of this study was that, in addition to finding many new isotopes and measuring their half-lives, we also obtained improved half-life values for several other isotopes. These new data have been reported previously[5,6], together with comparisons between the measured data and the predictions of two

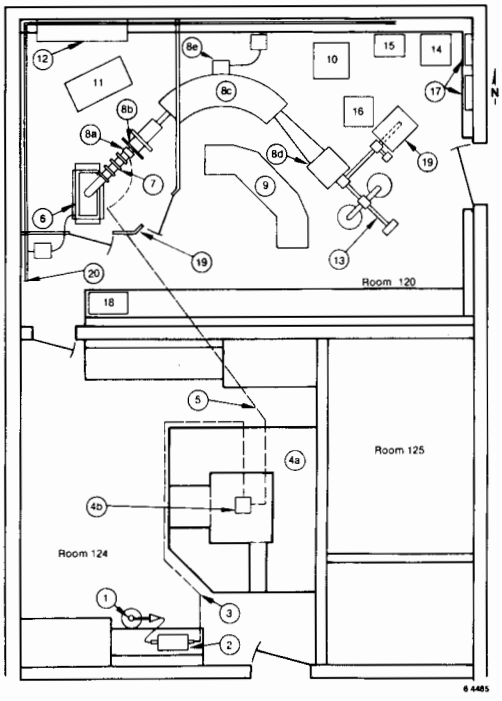

Fig. 1 Floor plan layout of the INEL ISOL facility. The two new collection tape transport lines are labeled as: (13) for coincidence spectroscopy and, (19) for total absorption gamma spectroscopy.

Table 1. Measured half-life values and their comparison to theory.

Z	Isotope	Experimental	Theoretical			R = (Observed/Predicted)		
		Present Work (sec)	Tachibana et al.(9)	Takahashi et al.(7)	Klapdor et al.(8)	Tachibana et al.(9)	Takahashi et al.(7)	Klapdor et al.(8)
59	^{151}Pr	18.90 ± 0.07	31.74	34		0.60	0.56	
	^{152}Pr	3.7 ± 0.2	12.66	13	3.93	0.29	0.28	0.94
	^{153}Pr	4.28 ± 0.11	8.05	13	2.81	0.53	0.33	1.53
60	^{153}Nd	28.9 ± 0.4	122.7	58		0.24	0.50	
	^{154}Nd	25.9 ± 0.2	32.65	77		0.79	0.34	
	^{155}Nd	8.9 ± 0.2	17.35	18	15.3	0.51	0.49	0.58
	^{156}Nd	5.47 ± 0.11	6.31	21	8.36	0.87	0.26	0.65
61	^{153}Pm	315 ± 1	756.1	229		0.42	1.38	
	^{155}Pm	41.5 ± 0.2	143.5	74	21.2	0.29	0.56	1.96
	^{156}Pm	26.70 ± 0.10	41.32	25	8.39	0.65	1.07	3.18
	^{157}Pm	10.50 ± 0.12	29.55	25	4.68	0.36	0.42	2.24
	^{158}Pm	4.8 ± 0.5	13.60	10	2.31	0.35	0.48	2.08
62	^{157}Sm	483 ± 2	923.4	110		0.52	5.3	
	^{158}Sm	318 ± 2	239.8	390		1.33	0.82	
	^{159}Sm	11.37 ± 0.15	91.02	37	10.1	0.12	0.31	1.13
	^{160}Sm	9.6 ± 0.3	18.63	74	6.06	0.52	0.13	1.58
63	^{161}Eu	24 ± 4	112.9	64	24.8	0.21	0.38	0.97
	^{162}Eu	10.6 ± 1.0	38.11	20	9.35	0.28	0.53	1.13
64	^{164}Gd	45 ± 3	70.82	229	31.8	0.64	0.20	1.42

existing models; namely that of Takahashi et al.[7] based on the gross theory of beta decay and that of Klapdor et al.[8] based on a microscopic model employing a description of the distribution of beta strength. Since these reports Tachibana et al.[9] have published half-life predictions based upon an improved gross theory of beta decay. In their model, improvements to the gross theory were made (1) to take into account pairing, (2) to obtain better agreement with the sum rule and, (3) to modify the description of the single-particle strength functions for Gamow-Teller and first forbidden transitions. It is thus of interest to see how these new half-life predictions compare to the earlier predictions and to the experimental data. This comparison is shown in Table 1. Disappointingly, we note that there appears to be no systematic improvement in predicted half-life values using the improved gross theory. The new model[9] still appears to systematically overestimate the measured half-life values, as did the older gross theory model.[7]

Total Beta-Decay Energies (Q_β)

Measurements of beta end-point energies to known levels in neutron-rich nuclides have provided a convenient method of deriving Q_β values, that is mass differences between neighboring isobars. Since nuclide masses close to the line of beta stability are generally known with good accuracy, such measurements allow nuclide masses in isoboric chains to be determined out to the limits of practical neutron-rich nuclide production, e.g., in fission. Extension of the region of know nuclide masses is an essential input to ongoing theoretical efforts to further refine and develop models to predict nuclide masses even further from stability.

As one moves further off the line of beta stability, nuclei have higher Q_β values and more complex beta-decay schemes. Thus, in order to reliably utilize beta end-point energies to obtain Q_β values, it is practically mandated that such end-point values be derived from beta spectra measured in coincidence with the corresponding de-exciting gamma rays.

We are presently developing such a beta-gamma coincidence system at the INEL ISOL facility. In order to allow for measurement of beta end-point energies of up to ~10 MeV, a 200-mm^2 x 10-mm intrinsic planar Ge detector was selected for beta spectral measurements (e.g., see Ref. 10). Although a Ge detector is also sensitive to gammas, we note that the gamma rays observed under the imposed coincidence conditions will have energies less than that of the beta end point. Because of its gamma sensitivity though, accurate energy calibrations can be made quite conveniently using gamma-lines.

In our detector arrangement, the cryostat housing is mounted into the vacuum system of the ISOL facility and the beta sources collected on the transport tape are viewed through a 50 μm Be window. Based upon experiments using ^{137}Cs and ^{207}Bi internal conversion electron sources, we obtained values for the electron energy losses in the Be window plus detector dead layer of 27.0, 24.6 and 23.0 keV at electron energies of 481.7, 624.2 and 975.6 keV, respectively, with the electron peaks being broadened to ~10.5 keV (FWHM).

Table 2. Q_β values deduced from beta end-points

Isotope	Q_β Value (keV)	
	This Work	1983 Mass Evaluation[11]
^{133}Xe	429[a]	427
	416[b]	
^{32}P	1703	1710.3
^{90}Y	2279	2281.5
^{144}Pr	2988	2997

[a] Uncorrected for β + 81-keV gamma summing
[b] Corrected for β + 81-keV gamma summing

Initial tests of this Ge spectrometer have been made using beta emitters with simple decay schemes. Using only the raw counting data without

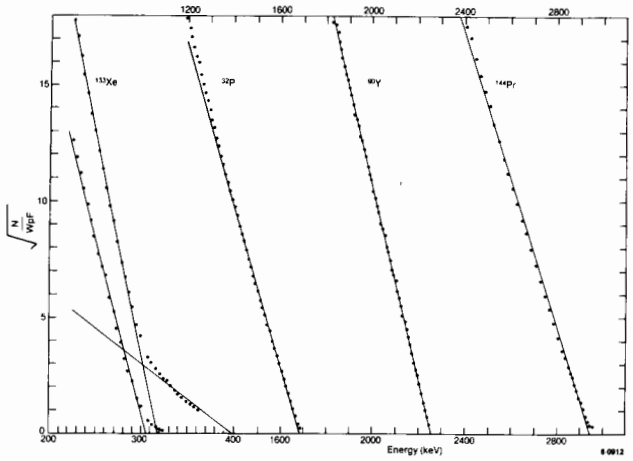

Fig. 2 Fermi-Kurie plots of the beta end-point energy regions of ^{133}Xe, ^{32}P, ^{88}Y and ^{144}Pr.

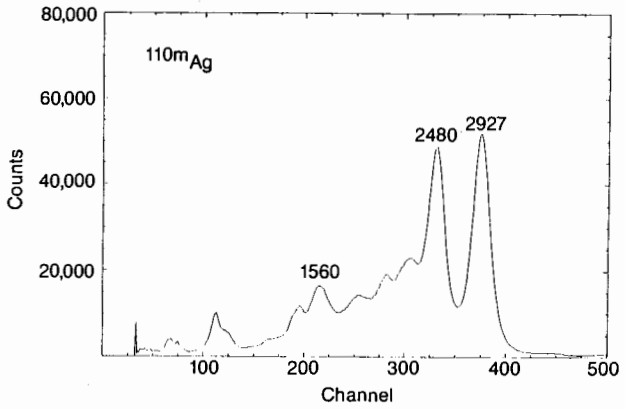

Fig. 3 Total absorption gamma spectrum of ^{110m}Ag. The small peaks, including the one at 1560 keV, below the sum peaks at 2480 and 2927 keV result from escape of one or more components of a cascade.

further correction for detector response, we obtain the Fermi-Kurie plots of the end-point regions shown in Fig. 2. The Q_β values derived from these data are shown in Table 2 and compare quite well with values obtained from the 1983 Mass Evaluation[11], with agreement being <20 keV.

Beta Strength Functions

For nuclei with large values of Q_β, where the density of final states at the higher level energies is large, a convenient way of describing the beta-decay process is to treat averages over large numbers of the final states using the concept of a beta strength function, $S_\beta(E)$[12]. Since the reduce beta-transition probability is normally defined in terms of a reciprocal ft value, it is convenient to also define $S_\beta(E)$ in the same manner. Thus, we have

$$S_\beta(E) = \frac{b(E)}{ft} = D^{-1} \sum_i B_i(E)$$

where; b(E) is the absolute beta decay branching intensity per unit energy; f is the statistical rate function; t is the beta-decay half-life; D is a constant (~6170 s); and, $B_i(E)$ is the reduced transition probability for each state populated in the unit energy interval. An understanding of the behavior of the distribution of beta strength in nuclei accessable to measurement is then clearly a prerequisit for more reliable prediction of beta-decay properties of nuclei further from stability. Such predictions must be made for example in nuclear astrophysical calculations involving the r-process.

A direct measurement of the distribution of beta strength is most straightforwardly accomplished using total absorption gamma spectrometry (e.g., see Refs. 13 and 14). An advantage of using this "direct" approach, apart from its inherent simplicity, is that it avoids the so-called "Pandemonium" problem (errors in deducted beta intensities due to nonobservation of a significent fraction of the emitted gamma radiation in complex decay schemes)[1,2].

A total absorption gamma spectrometer has very recently been installed at the INEL ISOL facility for beta-strength function measurements. The spectrometer consists of a large NaI(Tl) scintillator, with dimensions 25.4-cm diameter x 30.5-cm long and with a 5.1-cm diameter x 20.3-cm long axial well. This scintillator has excellent resolution, being 7.1% at 661-keV and ~3.5% at 6.8-MeV. The NaI(Tl) detector has been installed in a shielded cave at the ISOL facility and a new ion collection station and tape transport line has been developed (see Fig. 1) to allow for rapid movement of the mass-separated isotope samples into the well of the scintillator.

The use of total absorption spectrometry to obtain strength function distributions for β^- emitters is unfortunately not entirely straightforward in that a number of sometimes contradictory experimental requirements must be met. These requirements can be summarized as, (1) close to total gamma absorption in 4π geometry, (2) no detection of the betas in the scintillator, (3) no gamma absorption external to the scintillator (which is in clear contradiction with (2)), and (4) total-absorption-peak efficiencies which are independent of the decay paths from the beta-fed excited states. Thus, a simple arrangement employing a beta absorber surrounding the source, thick enough to stop 8 MeV betas provides too much gamma absorption. Instead, this need for minimal external gamma absorption mandates the use of a beta-gamma coincidence arrangement with the beta detector located in "poor" geometry (detection solid angle <10%) relative to the source. Such an arrangement unfortunately generates other sets of problems, arising from beta backscattering and the detection of internal conversion electrons and gammas in the beta detector, each of which can give rise to "false" coincident events. The design process then becomes one of evaluating the importance of each extraneous effect, minimizing it with design compromises, and estimating its effect in the selected design.

In the spectrometer configuration being tested, a 300-mm^2 x 1000-μm Si detector is used for coincident β^- detection. With this thickness detector, an energy discrimination requirement can be imposed to significantly reduce false coincident events resulting from detection of internal conversion electrons and low energy gamma rays. Initial tests of the system to evaluate its performance as a "total" absorption gamma spectrometer have used sources with simple two-and three-step cascades. In Fig. 3 we show the measured sum spectrum obtained from a somewhat more complex decay, ^{110m}Ag. The sum peaks at the two principal levels populated in this decay, at 2480 and 2927 keV, clearly dominate and show more complete summing than that reported in Ref. 14. Measurements of fission product isotopes are currently being made on-line with the ISOL facility.

Table 3. Delayed-neutron spectral intensities (relative) for the Rb and Cs precursor isotopes.

Neutron Energy Range (keV)	Relative Neutron Intensity							
	^{93}Rb	^{94}Rb	^{95}Rb	^{96}Rb	^{97}Rb	^{143}Cs	^{144}Cs	^{145}Cs
E_c[a] - 11.6		1.0	1.4	0.7	0.7	0.3	0.5	0.8
11.6- 19.0	1.0	1.7	8.4	1.4	0.9	2.6	2.7	1.6
19.0- 31.1	1.4	2.5	3.0	2.4	1.7	6.1	5.2	2.9
31.1- 45.0	1.1	2.0	1.3	2.9	1.7	3.3	4.9	3.2
45.0- 65.2	1.9	2.4	1.8	4.2	2.3	4.0	6.8	4.6
65.2- 83.5	2.1	2.5	2.2	5.0	3.7	5.8	5.9	4.8
83.5- 106.8	4.4	3.5	4.2	5.6	4.2	7.5	7.1	6.4
106.8- 136.7	6.3	5.8	3.6	6.3	4.5	8.6	8.7	7.1
136.7- 175.6	6.6	7.4	5.1	8.7	7.0	11.0	8.9	8.6
175.6- 224.1	8.5	8.8	7.3	9.6	6.9	12.9	9.4	8.8
224.1- 286.8	13.4	8.9	7.3	10.9	9.6	11.3	8.6	8.6
286.8- 367.2	13.4	12.3	7.2	12.2	9.6	9.8	7.7	8.8
367.2- 470.0	13.3	11.5	10.5	8.3	10.9	5.4	7.1	9.1
470.0- 601.7	12.1	10.3	13.8	7.4	11.9	4.6	5.9	8.9
601.7- 770.2	9.7	8.9	10.8	6.1	11.4	3.4	5.2	7.3
770.2- 985.9	3.5	5.9	5.8	4.0	7.0	2.0	3.1	4.6
985.9- 1262.0	1.0	2.9	3.2	2.5	3.5	1.0	1.7	2.5
1262.0- 1615.5	0.3	1.2	1.9	1.4	1.8	0.2	0.6	1.1
1615.5 - E^b	0.02	0.4	1.0	0.5	0.6	0.001	0.1	0.3
2068.0- 2647.2		0.04	0.18					
2647.2- 3150			0.02					

[a]Values of E_c, for $^{93-97}$Rb and $^{143-145}$Cs are 13.8, 8.0, 7.1, 8.0, 7.1, 10.2, 10.2 and 7.8 keV.
[b]Values of E are 1971.3 keV for ^{93}Rb and ^{96}Rb, 2017.0 keV for ^{97}Rb, and 2068.0 keV for ^{94}Rb and ^{95}Rb.

Delayed Neutron Energy Spectra

Experiments are being conducted using the TRISTAN ISOL facility at BNL to measure delayed-neutron energy spectra for those fission-product precursors most important to describing the kinetic behavior of nuclear reactors (particularly fast reactors). Recognizing that it is the lower energy delayed neutrons, if they are present with significant intensities, which would have the greatest effect on kinetic behavior, these experiments have principally used gas-filled proton-recoil proportional counters, because of their good energy resolution at low neutron energies and their insensitivity to thermal neutrons.

Details of the measurements are given in Refs. 15 and 16. The detectors being used in this work include a 2.6 Atm. H_2-gas counter (with PSD), 2.6 Atm. and 5.3 Atm. CH_4-gas filled proportional counters and a 5.1 cm x 1.27 cm BC-501 liquid scintillation detector (with PSD). Using these detectors we obtain good quality neutron spectral data over a wide energy range, from ~10 keV up to ~2 MeV, with discreet line structure being observed up to several hundred keV. In Table 3, measured delayed-neutron spectral-intensity distributions of $^{93-97}$Rb and $^{143-145}$Cs are summarized and compared (with course energy-bin widths ranging from ~0.5 to ~0.25 lethargy units and with the total intensity for each precuror being set over the total energy range measured to 100.)

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REFERENCES

1. C.W. Reich, Nuclear Data for Basic and Applied Science (Gordon and Breach, NY, 1986), p.647
2. C.W. Reich, Nuclei Off the Line of Stability, ACS Symposium Series 324 (1986) p.94
3. G.J. Mathews, NEANDC Specialists Meeting on Yields and Decay Data of Fission Product Nuclides, BNL-51778 (1983) p.485
4. R.A. Anderl et al., Nucl. Instrum. Methods B26, 333 (1987)
5. R.C. Greenwood et al., Phys. Rev. C35, 1965 (1987)
6. R.C. Greenwood, et al., Nuclei Far From Stability, AIP Conf. Proc. 164 (1988) p.782
7. K. Takahashi et al., At. Data Nucl. Data Tables 12, 101 (1973)
8. H.V. Klapdor et al., At. Data Nucl. Data Tables 31, 81 (1984)
9. T. Tachibana et al., Nuclei Far From Stability, AIP Conf. Proc. 164 (1988) p.614; and, private communication
10. T.A. Girard and F.T. Avignone III, Nucl. Instrum. Methods 154, 199 (1978)
11. A.H. Wapstra et al., Nucl. Phys. A432, 1 (1985)
12. P.G. Hansen, Adv. Nucl. Phys. 7, 159 (1973)
13. C.L. Duke et al., Nucl. Phys. A151, 609 (1970)
14. A.A. Bykov et al., Izvest. Akad. Nauk SSSR Ser. Fiz. 44, 918 (1980)
15. R.C. Greenwood and A.J. Caffrey, Nucl. Sci. Eng. 91, 305 (1985)
16. R.C. Greenwood and A.J. Caffrey, in Proc. Specialists Meeting on Delayed Neutron Properties, Univ. Birmingham Report (1987)