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Precision Mass Measurements of Heavy ^{252}Cf Fission Fragments Near the r-process Path

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The path of the astrophysical r process and the resulting elemental distribution are dependent on the neutron-separation energies of species far from stability. The mass models used to extrapolate these energies have increasing uncertainties with neutron number. Therefore, precision mass measurements of species near the r-process path are vital to generate more precise predictions of elemental abundances produced by this explosive nucleosynthetic process. As part of an ongoing program, the Canadian Penning Trap mass spectrometer at Argonne National Laboratory is measuring the masses of fission products from a 150-μCi ^{252}Cf source placed inside a new large-volume He gas catcher. Within the region investigated, new precision mass measurements have been made closer to the r-process path than have previously been published, with precisions near 15 keV/c^2. Presented measurements include isotopic chains of Pr, Nd, Pm, Sm, Eu and Gd to N=96, 97, 98, 99, 98, and 99, respectively, and our results differ from the AME03 [1] by up to 325 keV/c^2. Work will continue with the current fission source until 2009, when measurements of many more neutron-rich isotopes will be made at the CARIBU upgrade to the ATLAS accelerator at ANL.

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1. Introduction

The path of the rapid neutron capture process (r process) lies largely outside the limit of known masses in the region of high neutron excess. This poses difficulties for calculations of nuclear abundances produced by the *r* process and the determination of the exact path that it takes. Yields of species along the path depend exponentially on the neutron separation energy (S_n) as in the Saha equation [2]:

$$\frac{Y(Z,A+1)}{Y(Z,A)} = n_n \left(\frac{2\pi\hbar^2}{m_u kT}\right)^{3/2} \left(\frac{A+1}{A}\right)^{3/2} \frac{G(Z,A+1)}{2G(Z,A)} exp\left[\frac{S_n(Z,A+1)}{kT}\right]$$

Measurements of the masses of these species from which S_n can be determined are critical to a full understanding of this process.

The Canadian Penning Trap (CPT) mass spectrometer is undertaking a series of precision measurements targeted at pushing the limit of known masses yet closer to the *r*-process path. This paper is a report on the CPT system—which has seen several recent upgrades—as well as the progress of these measurements.

2. CPT Equipment

Major upgrades have been made to the CPT system in the two years between the group's previous fission fragment measurements and this work. A significantly larger gas catcher for stopping ions is now in operation on the second of two beam lines. Within it, a stronger ^{252}Cf source is supplying higher yields for the study of more neutron-rich species than were previously available. A new isobar separator, a gas-filled Penning trap, is online providing better rejection of contaminant ions. A brief description of the apparatus follows, and is more elaborately depicted in [4].

The nuclei studied are produced from spontaneous fission in a $\sim$150 μCi-^{252}Cf source inside the gas catcher (which is also used for collecting products from fusion-evaporation reactions from the ATLAS accelerator, as in [4]). The fission products are slowed by a gold foil and helium gas at a pressure of $\sim$80 mbar, and guided out of the gas catcher by a combination of DC and RF fields and gas flow. Next, an RFQ ion guide conveys them to a small potential well, where they are cooled by gas and accumulated for injection to the rest of the system.

After ejection from the RFQ, the bunch of ions travels through a beam line to the isobar separator for purification. In this gas-filled Penning trap [7], the ions are subjected to dipole and mass-selective quadrupole RF excitations and are purified with a mass resolution of 1 part in 5,000. A third excitation is applied to break up any molecular activity. Once cooled, the purified ions are ejected toward another gas-filled linear RFQ trap for final staging before transfer to the precision Penning trap for measurement.

In a Penning trap, ions are confined to one axis by a uniform magnetic field, and along that axis by a harmonic electrostatic potential. The cyclotron motion, with frequency ω_c, is split by the electric field into a fast "reduced cyclotron" motion and a slow magnetron motion with frequencies "ω_+" and "ω_-", respectively. An application of a quadrupole RF field at ω_c couples the motions.

The CPT is inside a highly stable 5.9 T magnetic field created by a superconducting solenoid. The ring electrode of the trap is split into quadrants to allow for the application of quadrupole

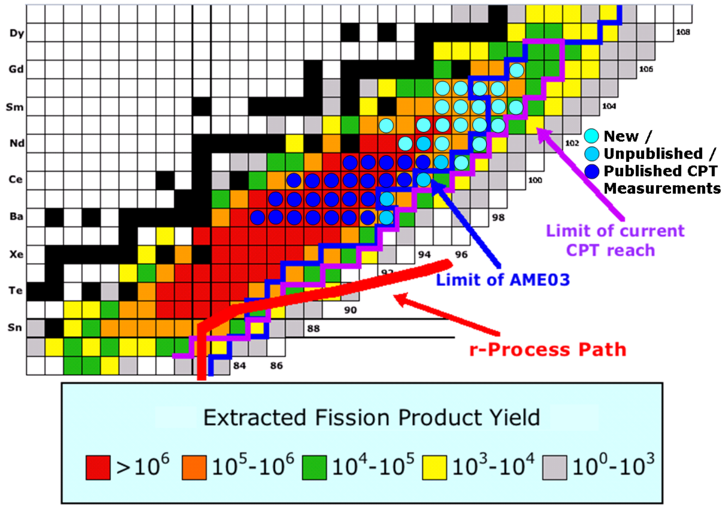

Figure 1: The target region of these measurements. Nuclides with circles on them indicate CPT measurements, with those reported here colored lightly, older not yet published measurements colored moderately, and published measurements [6] darkly. The red line indicates the approximate path of the *r* process. Nuclide coloration reflects ^{252}Cf spontaneous fission yields in counts per second per Curie of ^{252}Cf.

excitations, and small holes are in the centers of the end cap electrodes for capture and ejection of the ions. Additional correction electrodes compensate for electric field imperfections due to the holes and finite extent of the trap.

In the CPT, ions are subjected to a sequence of pulses and excitations. When a batch of ions is captured in the trap the remaining contaminants are removed by driving their ω_+ motions, then the purified ions are positioned in an orbit at the magnetron frequency by a brief ω_- dipole excitation. Each batch is then subjected to a quadrupole field at a frequency near the ω_c expected for the species of interest. Next, the ions are ejected from the trap, and the fringe magnetic field converts their orbital motion to linear motion down the beam line. A microchannel plate detector at the end of the line measures the time of flight (TOF) of the ions. If the ions' motion inside the trap is converted from the slow magnetron motion to the fast cyclotron motion, their TOF will be reduced. By cycling through different applied frequencies for each bunch, a TOF spectrum is produced. The minimum of this spectrum corresponds to the cyclotron frequency of the ions, and thus a measurement of the mass to charge ratio [3].

3. Measurements thus far

After a series of successful measurements [4, 5] of nuclei made on-line with the ATLAS accelerator used all the new components of the CPT system, a still-ongoing program of fission fragment measurements restarted in April 2008. This program resumes the group's previous program of fission fragment measurements [6]. The region of interest, shown in Fig. 1, was selected for several reasons: the unique capability of a ^{252}Cf source to produce masses greater than those produced from Uranium fission, the abundance of 2^+ ions above the xenon electron shell closure, and the proximity of the *r*-process path.

During four weeks from April to July 2008, the masses of 20 different nuclides were measured. The target precision was one part in 10^7, or about 15 keV/c^2, which is sufficient precision to address many astrophysical questions. Ions used were in the 2^+ charge state, doubling the cyclotron frequency and thus the precision of the mass measurement. Each species was subjected to a quadrupole excitation length of 200, 500, or 1,000 ms, depending on factors such as purity,

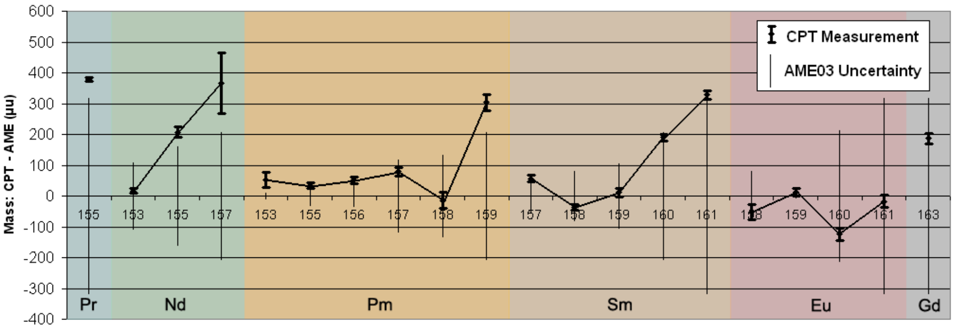

Figure 2: Results of the CPT mass measurements grouped by element and in order of increasing A, and shown as the difference from the AME03 [1] values. Note the trend upwards from the AME03 masses, particularly in the highest A species where there are no β-decay studies.

yield, and lifetime. Longer excitations provide higher precision, as the resolving power of the TOF spectrum is Fourier limited. The time taken to collect data for each species varied from a few hours to over a day. After each group of measurements, calibration ω_c spectra were taken with singly-charged $^{12}C_6{}^1H_4$, ^{80}Kr, and ^{86}Kr. The ratio of the cyclotron frequencies of the studied ions to the calibrant ions is the inverse of the ratio of their masses over charge. Thus the mass of the neutral atom is given by:

$$m = 2\frac{\omega_{cal}}{\omega_c}(m_{cal} - m_{e^-}) + 2m_{e^-}$$

where the factors of 2 come from the difference in charge states between the studied and calibrant ions.

Four of these masses have never before been measured by any means (^{155}Pr, ^{157}Nd, ^{159}Pm, and ^{161}Sm), and most of the rest were improved by a typical factor of 5–10, having only been previously measured via β-decay studies. As shown in Fig. 2, the species nearer stability (lower A) agree well with the 2003 Atomic Mass Evaltuation (AME03) [1]. However, farther from stability our measured masses trend heavier than the AME03 extrapolations, which results in lower S_{2n} values, as shown in Fig 3. This continues the trend seen in our previous work [6]. If yet higher

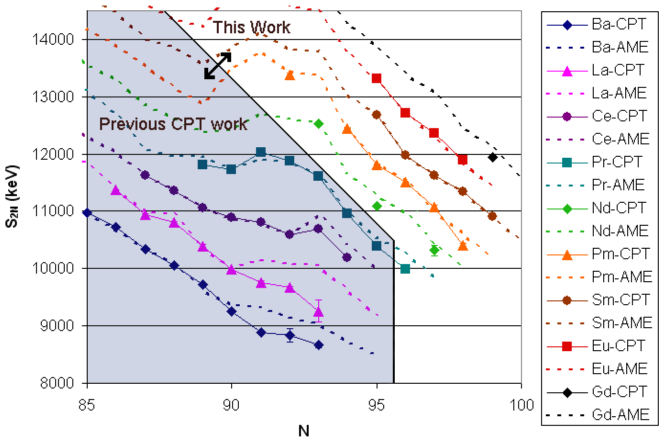

Figure 3: The two-neutron separation energies (S_{2n}) of measured species plotted with the S_{2n} from the AME03 [1]. The shaded area is where CPT's previous measurements lie [6], with new measurements above and to the right. At high mass the measured values dip below AME03 for most elements.

N species follow this trend then both the *r*-process path and the neutron drip line will be closer to stability than previously thought.

These measurements will continue through 2008 and move to include the rest of the heavy fission peak, where 20 to 25 species are accessible and lacking precision measurements. The TOF detector is being upgraded to a channeltron, which is expected to increase the efficiency of the system by a factor of three. Early in 2009, the CPT will be moved to the CARIBU facility currently under construction at Argonne to continue the push toward the *r*-process path.

4. Future: CARIBU

The Californium Rare Isotope Breeder Upgrade (CARIBU) currently under construction at the ATLAS accelerator at ANL will provide much higher yields of neutron-rich isotopes than are currently available for the CPT. The front end of CARIBU is an even larger version of the gas catcher and ^{252}Cf source used for the measurements presented here, as well as a demonstration of technology for use in the Facility for Rare Isotope Beams that ANL has proposed. The upgrade will provide fission fragments both for low energy experiments such as the CPT, and Coulomb barrier energy experiments by serving as a source for ATLAS.

CARIBU will come online in early 2009 with a 80 mCi ^{252}Cf source which will later be upgraded to 1,000 mCi. A new isobar separator will be part of this system, providing a mass resolution of one part in 20,000. When the CPT is online at CARIBU, the 7,000-fold increase in fission product yields will allow for well over 100 previously unknown masses to be measured to high precision. The data that will come from the CPT at CARIBU will vastly improve our knowledge of the *r*-process behavior, as many of the species to be measured lie directly on its expected path.

PoS(NIC X)150

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