

## NEW NEUTRON-DEFICIENT ISOTOPES OF BARIUM AND RARE-EARTH ELEMENTS

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This paper deals with an investigation of the short-lived neutron-deficient isotopes of barium and rare-earth elements. By using the BEMS-2 isotope separator on a heavy ion beam, we succeeded in producing 19 new isotopes with mass numbers ranging from 117 to 138. Five of these ( $^{117}\text{Ba}$ ,  $^{129,131}\text{Nd}$  and  $^{133,135}\text{Sm}$ ) turned out to be delayed proton emitters. The  $\beta$ -decay probabilities for the new isotopes have been analyzed in terms of the  $\beta$ -strength function. An analysis of the proton spectrum shape has been performed using the statistical model for delayed proton emission.

### 1. Experimental Technique

The experiments were carried out using an external beam from the JINR Laboratory of Nuclear Reactions U-300 Heavy Ion Cyclotron. Targets made of the enriched isotopes  $^{92}\text{Mo}$ ,  $^{96}\text{Ru}$ ,  $^{102}\text{Pd}$ ,  $^{106}\text{Cd}$  and  $^{112}\text{Sn}$  were bombarded with (180-190) MeV  $^{32}\text{S}^{5+}$  ions with the beam intensity of (1-3)  $\mu\text{A}$ . The targets were positioned in the close vicinity of the BEMS-2 on-line ion source<sup>1)</sup>. In these experiments, use was made of a high-temperature surface-ionization ion source with an operating temperature of up to 2700°K, described in detail in ref.<sup>2)</sup>. The efficiency of the isotope separator for rare-earth elements was 15 - 20 % under these operating conditions, the average ion hold-up time in the ion source being (5-10) sec. The ion source was designed to permit the use of two alternate targets with a possibility of replacing them without destroying the vacuum.

Through a 2 mm slit in the focal plane of the isotope separator isobars

with a certain mass passed. Behind the slit, a thin (1  $\mu\text{m}$ ) aluminium catcher foil glued onto a disk with holes was placed. The detecting system consisted of two Si(Au) proton counters with an energy resolution of  $\sim 40$  keV, a Ge(Li)  $\gamma$ -ray detector (with a sensitive volume of 23  $\text{cm}^3$  and energy resolution of about 5 keV for  $^{60}\text{Co}$ ), a Princeton Gamma-Tech. X-ray detector (with an active area of 25  $\text{mm}^2$ , a thickness of 5 mm and energy resolution of about 350 eV) and two plastic scintillation  $\beta$ -counters. The accumulated activity of the isobar chosen was placed from time to time between the proton counter and the  $\beta$ -counter, or between the Ge(Li) (or X-ray) detector and the second  $\beta$ -counter. The other Si(Au) proton detector was placed on-line with the separated isotope beam behind the catcher foil.

For the mass calibration of the isotope separator the  $\beta$ -counting rate was measured as a function of the magnetic rigidity. The curve obtained shows pronounced peaks corresponding to the detection of isobars with definite mass numbers (fig. 1). The assignment of the mass numbers to the peaks was made on the basis of the properties of the known radioactive isotopes produced in the same experiment.

### 2. Identification of New Isotopes.

In the synthesis of new isotopes, heavy ion reactions of the (HI,xn) type are widely used. For the identification of an isotope, the excitation function of the reaction leading to the formation of the final product are usually measured. However, this method becomes unsuitable if one goes to the very proton-rich isotopes of medium-weight elements because at a fixed excitation energy the (HI,xp,yn) and (HI, $\Delta$ ,xp,yn)

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reactions leading to isobars with given  $A$  but different  $Z$  values occur with comparable probabilities.

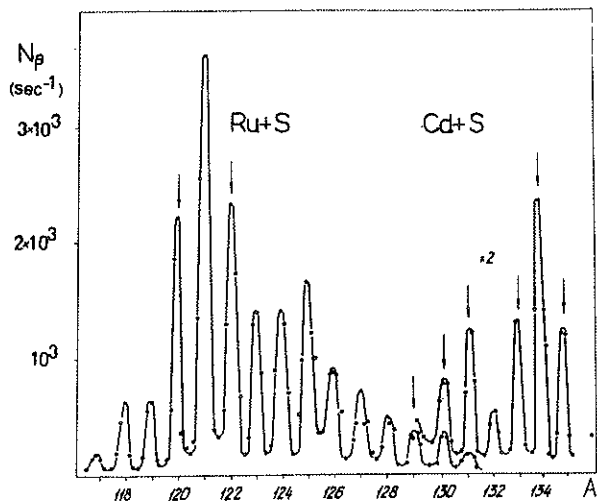


Fig. 1. The  $\beta$ -counting rate on the catcher foil in the focal plane measured as a function of the isobar mass number. Arrows show isobars for which new isotopes have been observed. The activity accumulation and counting times are equal to 13 sec.

In the present paper isotopes were mainly identified by measuring the  $\gamma$ -ray and X-ray spectra of the daughter nuclei formed as a result of the  $\beta^+$ -decay. In addition, the decay curves of the total  $\beta$ -activity of given isobars have been measured. To eliminate the contribution due to the decay of the low-lying isomeric states, the  $\gamma$ - and X-ray spectra were measured in coincidence with positrons (the threshold in the  $\beta$ -channel was set at a level of (0.5-1.0)MeV). All of the measurements were carried out with the electronics operated in the time-amplitude mode. As an example, fig. 2 shows the X-ray spectrum measured for isobars with  $A=133$ . Here one can clearly see the characteristic X-ray lines corresponding to Ce, Pr and Nd. The results are summarized in table 1.

Now we shall elucidate the case where isotopes were identified without measuring the characteristic X-radiation.

In the  $\gamma$ -spectrum for the  $A=120$  isobar, one observed the  $2^+ \rightarrow 0^+$  transition in  $^{120}\text{Xe}$  produced as a result of the

$\beta^+$ -decay of  $^{120}\text{Cs}$  ( $T_{1/2}=(62\pm 5)\text{sec}$ ). In addition, the 61 keV and 182 keV lines with half-lives of  $(32\pm 5)\text{sec}$  were also observable. These activities were attributed to  $^{120}\text{Ba}$ . Under the chosen operating conditions of the ion source<sup>2)</sup>, the separation efficiency for the heavier elements produced by nuclear reaction was depressed.

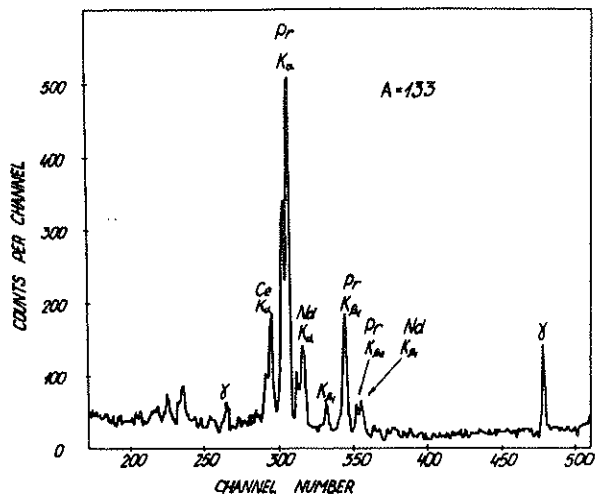


Fig. 2. The X-ray spectrum measured for isobars with  $A=133$  in the bombardment of  $^{106}\text{Cd}$  with 180 MeV  $^{32}\text{S}$  ions.

From ref.<sup>3)</sup> it is known of two isomers of  $^{122}\text{Cs}$ . The first one has a half-life of about 23 sec and the most probable spin and parity of  $2^+$ , while the second seems to be characterized by higher spin and a half-life of 4.2 min. In the  $\gamma$ -ray spectrum of the  $A=122$  isobar we observed only the  $2^+ \rightarrow 0^+$  transition in  $^{122}\text{Xe}$ , the intensity of this line decreasing with a half-life of  $(120\pm 20)\text{sec}$ . We attributed this half-life to  $^{122}\text{Ba}$ , whose decay leads to the population of the first isomer of  $^{122}\text{Cs}$ . Under the chosen operating conditions of the ion source the separation efficiency for the heavier elements was also depressed similarly to the previous case.

As one can see from the table, the isotopes  $^{117}\text{Ba}$ ,  $^{129}\text{Nd}$ ,  $^{131}\text{Nd}$ ,  $^{133}\text{Sm}$  and  $^{135}\text{Sm}$  are delayed proton emitters. Two of these isotopes,  $^{131}\text{Nd}$  and  $^{135}\text{Sm}$ , were identified by X-radiation. The rest of them were identified by the proton activity half-lives taking into account the existing empirical regularities for delayed proton emitters. All of the known proton

Table 1

The properties of the isotopes observed.

	A	$T_{1/2}$ , sec	Radiation measured	$\gamma$ -ray energy, keV	Reaction
Ba	117	$1.9 \pm 0.2$	P		$^{92}\text{Mo} + ^{32}\text{S}$
	120	$32 \pm 5$	$\gamma - \beta^+$	51, 182	$^{96}\text{Ru} + ^{32}\text{S}$
	122	$120 \pm 20$	$\beta^+, \gamma - \beta^+$		" "
Pr	129	$24 \pm 5$			$^{102}\text{Pd} + ^{32}\text{S}$
	130	$28 \pm 6$	$\beta^+, X$		$^{106}\text{Cd} + ^{32}\text{S}$
	131	$100 \pm 20$	$\beta^+, X$		" "
Nd	129	$5.9 \pm 0.6$	P		$^{102}\text{Pd} + ^{32}\text{S}$
	130	$28 \pm 3$	$\beta^+, X$		$^{106}\text{Cd} + ^{32}\text{S}$
	131	$24 \pm 3$	$\beta^+, P, X$		" "
	132	$105 \pm 10$	X		" "
	133	$70 \pm 10$	$\beta^+, X, \gamma$	63, 106, 166, 227, 251, 369	" "
Pm	132	$4 \pm 2$	X		" "
	133	$12 \pm 3$	X		" "
	134	$24 \pm 2$	$\beta^+, \gamma, X$	294, 460, 495, 632	" "
	135	$55 \pm 10$	$\gamma, X$	129, 199, 271, 364, 465	" "
Sm	133	$32 \pm 0.4$	P		" "
	134	$12 \pm 3$	X		" "
	135	$10 \pm 2$	P, X		" "
Eu	138	$35 \pm 6$	$\beta^+$		$^{112}\text{Sn} + ^{32}\text{S}$
		$1.5 \pm 0.4$	$\beta^+$		

emitters of medium-weight and heavy elements with odd mass numbers have even Z values. This indicates that the proton emission of the A=129 and A=133 isobars can, in principle, be related to  $^{129}\text{Sm}$  and  $^{133}\text{Gd}$  rather than  $^{129}\text{Nd}$  and  $^{133}\text{Sm}$ . However, we consider this conclusion unlikely since, as predicted by the gross theory of  $\beta^-$ -transitions, the  $^{129}\text{Sm}$  and  $^{133}\text{Gd}$  isotopes are expected to have half-lives by an order of magnitude shorter than those measured for the proton emission of the A=129 and A=133 isobars. As to the proton emission of the A=117 isobar, it is unambiguously related to  $^{117}\text{Ba}$ , since the reaction leading to the formation of  $^{117}\text{Ce}$  was energetically impossible in our experiments.

Fig. 3 shows the region of Z and N of the isotopic chart where the present investigations were carried out.

### 3. $\beta^+$ -Decay Strength Functions.

The systematics of the average values

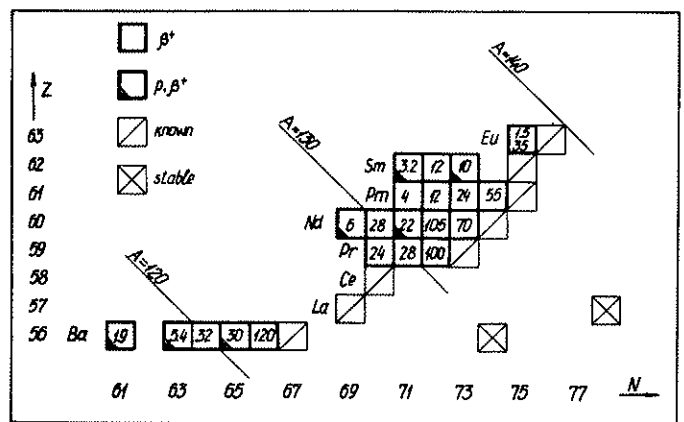


Fig. 3. Part of the isotopic chart showing the isotopes observed in the present investigation.

of the  $\beta^+$ -decay strength functions for Ba, Pr, Nd, Pm, Sm and Eu isotopes in the region of  $60 \leq N \leq 81$  is presented in fig. 4. The average values of the  $\beta^-$ -strength function  $\bar{S}_\beta$  was found using the relation

$$\bar{S}_\beta = [T_{1/2} \int_C^{Q_0} F(Z, Q_0 - E) dE]^{-1}$$

where  $F(Z, Q_0 - E)$  is the statistical rate function for the  $\beta^+$ -decay and electron capture,  $Q_0$  is the total energy of the K-capture, and  $E$  is the excitation energy of the daughter nucleus. The  $Q_0$  values were taken from ref.<sup>5)</sup>. The cutoffs  $C$  taking into account the pairing effect were chosen to be equal to 0 for doubly-odd daughter nuclei,  $14.4 A^{-1/2}$  MeV for nuclei with odd  $A$  and  $28.8 A^{-1/2}$  MeV for doubly-even nuclei<sup>6)</sup>. The isotopes shown by closed marks are those produced in the present work, while the rest of the data are taken from ref.<sup>7)</sup>. The dashed curve presents the average dependence of  $\bar{S}_\beta$  on the mass number according to the gross theory of K.Takahashi and M.Yamada. The calculation was made in ref.<sup>6)</sup> for an  $\beta$ -decay energy of 6 MeV and an excitation energy of 2 MeV. An increase in the  $\bar{S}_\beta$  value for  $N=80-81$  is apparently due to the effect of the  $d_{5/2}^p \rightarrow d_{3/2}^n$  transition. As one moves to the smaller values of  $N$ ,  $\bar{S}_\beta$  shows a weak dependence on  $N$  and  $Z$  thus justifying the statistical approach to the description of the  $\beta$ -decay at a high energy. It is noteworthy that in calculating  $\bar{S}_\beta$  the  $Q_0$  values taken from ref.<sup>5)</sup> were used. A 0.5 MeV variation in  $Q_0$  leads to a 50 % change in  $\bar{S}_\beta$ .

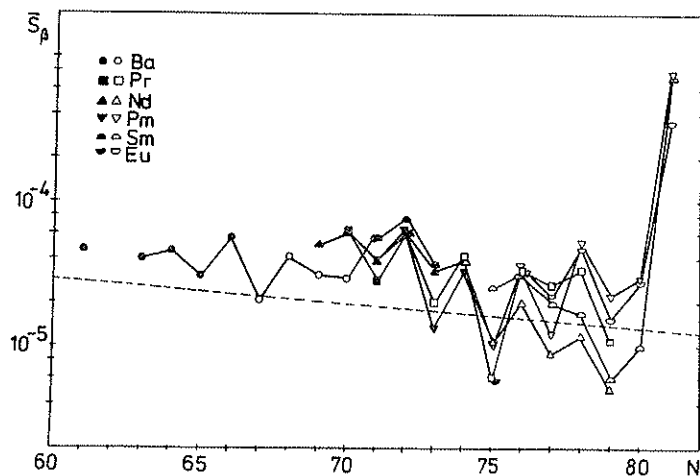


Fig. 4. Values of the averaged strength function ( $\text{MeV}^{-1} \text{sec}^{-1}$ ) of the  $\beta^+$ -EC-decay in the region of 61  $N$  81 for Ba, Pr, Nd, Pm, Sm and Eu isotopes.

#### 4. Delayed Proton Emitters.

The isotopes  $^{129,131}\text{Nd}$  and  $^{133,135}\text{Sm}$  are the first delayed proton emitters observed in the rare-earth region. The proton spectra of  $^{117}\text{Ba}$ ,  $^{133}\text{Sm}$  and  $^{135}\text{Sm}$  are presented in figs. 5-7.

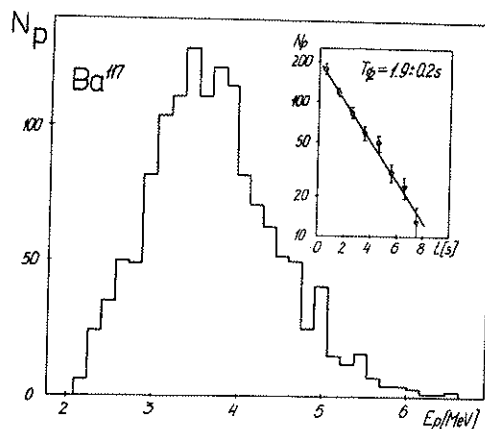


Fig. 5. The delayed proton spectrum and decay curve for  $^{117}\text{Ba}$ .

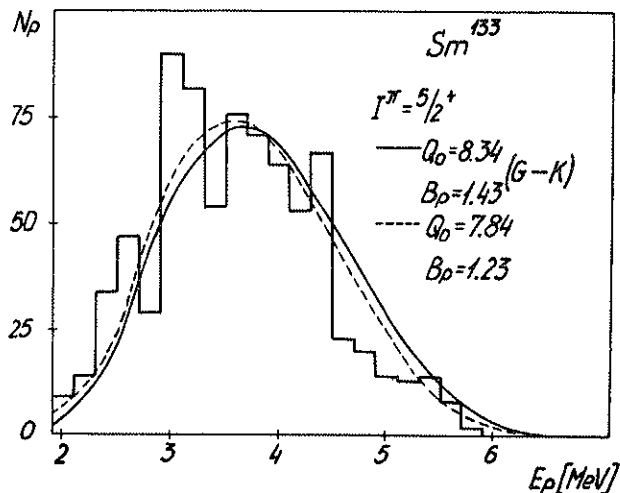


Fig. 6. Comparison of the delayed proton spectrum for  $^{133}\text{Sm}$  (histogram) with the calculation using the statistical model. The parameters used in the calculation are indicated in the figure. The  $Q_0$  and  $B_p$  values are given in MeV.

The experimental spectra of samarium isotopes are compared with those calculated on the basis of the statistical model of delayed proton emission<sup>8,9)</sup>. In this model, the averaged spectrum is described by the formula

$$I(E_p) = \sum_{if} F \frac{\langle \mu_i^2 \rangle}{D_{if}} \left\langle \frac{\Gamma_p}{\Gamma} \right\rangle_{if}$$

The summing-up is made over all initial (i) and final (f) states admissible by the selection rules. In the calculation of the relative proton width  $\langle \frac{\Gamma_p}{\Gamma} \rangle$  use was made of proton transmission coefficients from ref.<sup>10)</sup> and  $\gamma$ -ray transition widths from ref.<sup>11)</sup>. The quantity  $\langle \mu_i^2 \rangle / D_{if} = S_{\beta i}$  is the strength function for the  $\beta$ -transition of nucleons with spin j to the state with spin i and excitation energy  $E = E_p(A/A-1) + B_p + E_p$ .  $\langle \mu_i^2 \rangle$  is a  $\beta$ -matrix element squared and  $D_{if}$  is an averaged level spacing. The strength functions for the  $\beta$ -transition to states with different spins are correlated as follows

$$S_{\beta i} = g(j, i) S_{\beta}$$

where  $g(j, i)$  is a statistical weight factor. The  $\beta$ -strength function  $S_{\beta}$  was assumed to be constant. The numerical parameters of the calculation are  $Q_0 = B_p$  and  $B_p$ , where  $B_p$  is the proton binding energy in the daughter nucleus formed following the  $\beta$ -decay.

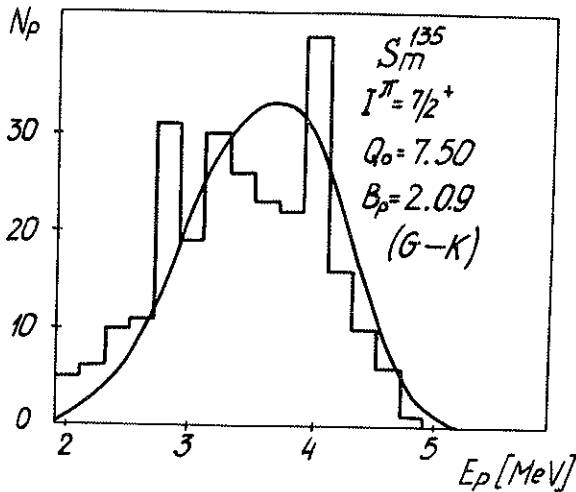


Fig. 7. The same as in fig. 6, for  $^{135}\text{Sm}$ .

The calculation performed shows a fairly good agreement between experimental and theoretical spectra obtained using the  $Q_0$  and  $B_p$  values predicted by the semi-empirical mass formula of Garvey and Kelson<sup>5)</sup>.

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