

days, one for a month, and two for six months. The samples were dissolved in warm nitric acid. Aliquots were evaporated with ruthenium metal carrier, and fused with a KOH-KNO<sub>3</sub> mixture to ensure exchange between fission product and carrier. The ruthenium was then converted to RuO<sub>4</sub> by periodate oxidation and purified by distillation and solvent extraction.

The Ba<sup>140</sup> activities in the 7-day and one-month irradiations were determined by the method of Glendenin and Steinberg,<sup>5</sup> modified by Cook.<sup>6</sup> The Cs<sup>137</sup> activities in the six-month irradiations were determined by J. G. Cuninghame and M. Sizeland, using a perchlorate separation followed by  $\beta$  counting.

From the saturation activities, the fission yields of Ru<sup>103</sup> and Ru<sup>106</sup> were calculated relative to Ba<sup>140</sup> for the seven-day and one-month irradiations. The fission yield of Ba<sup>140</sup> was taken to be 6.2 percent. The saturation activities for Ru<sup>106</sup> and Cs<sup>137</sup> were calculated for the 6-month irradiation, and assuming a fission yield of 6.15 percent<sup>7</sup> for Cs<sup>137</sup>, the fission yield of Ru<sup>106</sup> was found. The values obtained are given in Table I.

TABLE I. Fission yields of Ru<sup>103</sup> and Ru<sup>106</sup>.  
Summary of experimental results.

Irradiation period	7 days	1 month	6 months	
Standard	Ba <sup>140</sup>	Ba <sup>140</sup>	Cs <sup>137</sup>	Cs <sup>137</sup>
Fission yield Ru <sup>103</sup>	2.8	2.9	—	—
Fission yield Ru <sup>106</sup>	0.37	0.38	0.38	0.39

The probable error in the determination of the value for Ru<sup>103</sup> is  $\pm 5.7$  percent and for Ru<sup>106</sup>  $\pm 7.6$  percent. These errors are largely due to the uncertainties in the fission yields of Ba<sup>140</sup> and Cs<sup>137</sup> and the half-lives of Ru<sup>106</sup> and Cs<sup>137</sup>.

Thus the values for the percentage fission yields were found to be  $2.85 \pm 0.16$  for Ru<sup>103</sup> and  $0.38 \pm 0.03$  for Ru<sup>106</sup>. They are lower than Glendenin and Steinberg's values which were 3.7 and 0.52 for Ru<sup>103</sup> and Ru<sup>106</sup>. In both determinations the ratio of the Ru<sup>103</sup> and Ru<sup>106</sup> yields is nearly the same, being approximately seven.

I am grateful to Dr. J. M. Fletcher for suggesting this work and to him and Dr. G. B. Cook for most helpful discussions; also to the Director of the Atomic Energy Research Establishment, Harwell, for permission to publish this letter. Full details are being given in Atomic Energy Research Establishment Report C/R 1180.

<sup>1</sup> L. E. Glendenin and E. P. Steinberg, *Radiochemical Studies: The Fission Products* (McGraw-Hill Book Company, Inc., New York, 1951), Paper No. 103, National Nuclear Energy Series, Plutonium Project Record, Vol. 9, Div. IV.

<sup>2</sup> E. Kondaiah, *Phys. Rev.* **79**, 891 (1950).

<sup>3</sup> J. G. Mei, *Phys. Rev.* **79**, 429 (1950).

<sup>4</sup> J. M. Fletcher and J. Story, Harwell Atomic Energy Research Establishment Report C/R 1078 (unpublished).

<sup>5</sup> L. E. Glendenin and E. P. Steinberg, reference 1, Paper No. 288.

<sup>6</sup> G. B. Cook and H. Willis (unpublished).

<sup>7</sup> H. G. Thode, *Can. J. Phys.* **31**, 517 (1953).

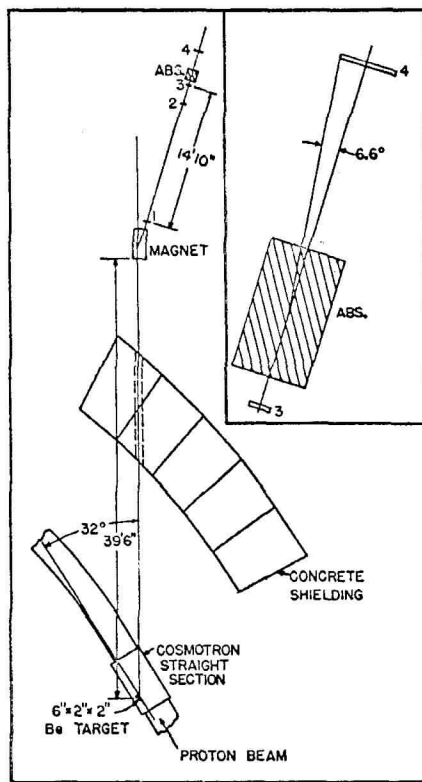


FIG. 1. Experimental arrangement.

The resolution of the analyzing magnet was measured using a current-carrying wire. The half-angle which the third defining counter subtended at the magnet was  $0.32^\circ$ , corresponding to a meson energy resolution of  $\pm 20$  Mev. Carbon and polyethylene absorbers were placed after the third counter, and at a variable distance behind them a large fourth counter measured the transmitted beam.

Fast plastic scintillators were used, the first three being  $2\frac{1}{2}$  in. and the fourth  $6\frac{1}{4}$  in. in diameter. Three- and four-fold coincidences were made in a Garwin-type<sup>2</sup> circuit, modified<sup>3</sup> in a manner permitting fast multifold coincidences. The resolving time of the circuit was about  $4 \times 10^{-9}$  sec.

The composition of the analyzed beam of negative particles entering the telescope was determined by careful analysis of the absorption of the beam in Cu and Pb. The only contamination observed was that of  $\mu$  mesons, which amounted to  $(2 \pm 2)$  percent and  $(5 \pm 3)$  percent for the 870- and 500-Mev beams, respectively. (These energies correspond to 840 and 470 Mev mean energy in the absorbers.)

Runs made with two different sets of absorbers and taken over a period of several months have given statistically consistent results. The carbon absorber was spaced out to fill the same physical volume as the polyethylene, and the two contained nearly equal numbers of carbon atoms. Most of the data were taken with 28.18 g/cm<sup>2</sup> of C and 32.65 g/cm<sup>2</sup> of CH<sub>2</sub>, which gave a hydrogen attenuation of approximately 7 percent. The C and CH<sub>2</sub> absorbers were frequently interchanged to eliminate the effects of any slight drifts in the equipment.

The 840-Mev measurements were made with two geometries. In the "poor" geometry, the fourth counter subtended at the center of the absorber a half-angle of  $6.6^\circ$ ; in the "good" geometry, a half-angle of  $2.1^\circ$ . The  $\pi^- - p$  cross sections measured with these two geometries, and corrected for the  $\mu$  contamination, were  $44 \pm 4$  mb and  $57 \pm 5$  mb, respectively. (All errors are standard deviations.) These values of  $\sigma$  must be corrected for two secondary

## The Total $\pi^- - p$ Cross Section at 840 and 470 Mev\*

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THE total  $\pi^- - p$  cross section has been measured at 840 Mev. Since this value appeared considerably higher than that measured at 450 Mev,<sup>1</sup> the measurement was repeated at the lower energy to confirm the difference with the same experimental arrangement and apparatus.

The experimental arrangement is shown in Fig. 1. A Be target, 6 in.  $\times$  2 in.  $\times$  2 in., was placed in a straight section of the Brookhaven cosmotron and bombarded by 2.2-Bev protons. Particles emitted from the target at an angle of  $32^\circ$  to the proton direction passed through a 3-in. diameter collimator in the concrete shielding wall.

This beam was then magnetically analyzed, and particles of the proper sign and momentum entered a threefold defining telescope.

effects which are different for the two geometries. The value of  $\sigma$  (poor) is low because some products of the  $\pi^- - p$  interaction will pass through the last counter. On the other hand,  $\sigma$  (good) is high because a few more mesons are multiply scattered out of the last counter by the  $\text{CH}_2$  than by the C attenuator, when the "good" geometry is used. We have assumed that 3 percent of the  $\pi^- - p$  interactions give a charged secondary traveling within  $6.6^\circ$  of the beam direction, and 0.3 percent within  $2.1^\circ$ . This corresponds to the condition that all the interactions are elastic scatterings with an isotropic distribution in the center-of-mass system, or to the condition that  $\frac{1}{3}$  of the interactions are elastic scatterings with a  $\cos^2\theta$  distribution and that the rest send no charged products into the last counter. From a detailed multiple Coulomb scattering calculation it was found that about 9 and 7 percent of the mesons were scattered out of the last counter by the  $\text{CH}_2$  and C absorbers, respectively, in the "good" geometry position, while no mesons were so lost in the "poor" geometry position. With these corrections the value of  $\sigma$  for both geometries becomes 47 mb. Thus the total  $\pi^- - p$  cross section is  $47 \pm 5$  mb, for a mean energy in the absorber of 840 Mev. The stated error includes an estimate of the uncertainties in the corrections.

We have repeated the above measurements with 500-Mev incident mesons using mainly the  $6.6^\circ$  geometry. With similar corrections, the total  $\pi^- - p$  cross section was found to be  $27 \pm 5$  mb for a mean pion energy of 470 Mev. This agrees well with the value of  $25 \pm 3$  mb at 450 Mev found previously at this laboratory.<sup>1</sup>

We find therefore a rise of approximately 20 mb in the total  $\pi^- - p$  cross section from 470 to 840 Mev. Since the cross section has been found to be  $34 \pm 3$  mb at 1.5 Bev,<sup>4</sup> and, very recently,  $47 \pm 4$  mb at 1.0 Bev,<sup>4</sup> a second peak in the neighborhood of 900 Mev is indicated. Further study of this energy region has been undertaken by Lindenbaum and Yuan, and Piccioni and Cool.

We should like to thank Dr. George B. Collins and Professor J. Steinberger for their important roles in the initial stages of this experiment, and the many members of the cosmotron group who made it possible.

\* This work was performed under the auspices of the U. S. Atomic Energy Commission.

<sup>1</sup> S. J. Lindenbaum and L. C. L. Yuan, Phys. Rev. (to be published).

<sup>2</sup> R. L. Garwin, Rev. Sci. Instr. 21, 569 (1950).

<sup>3</sup> Leon Madansky (private communication).

<sup>4</sup> Cool, Madansky, and Piccioni (private communication).

## Evidence for Subshell at $Z=96$

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THE evidence is decisive that major nuclear shells are completed at 82 protons and 126 neutrons (both represented by the nuclide  $\text{Pb}^{208}$ ) and these, along with major shells at 82 neutrons and certain lower nucleon numbers ( $N$  or  $Z=20, 28, 50$ ), are well explained by the strong spin-orbit coupling model of Mayer<sup>1</sup> and Haxel, Jensen, and Suess.<sup>2</sup> This model suggests the filling of quantum states at certain intermediate points, and there is an accumulating amount of evidence that such "subshells" are also discernible, for example, at  $Z=58^{3-5}$  and  $Z=64.$ <sup>6,7</sup>

The evidence from alpha radioactivity, both (1) the effect of the nuclear radius shrinkage on the relationship between energy and half-life and (2) the discontinuities in the plots of energy vs mass number at constant  $Z$ , gives a striking indication<sup>8</sup> of the closing of major shells at  $Z=82$  and  $N=126$ . Application of these sensitive criteria as tests for the much smaller "subshell" effects in the regions  $Z>82$  and  $N>126$  leads to some evidence for such a subshell at  $Z=96$  (curium).

Since it has been shown that the Gamow-Gurney-Condon type of formula for alpha decay applies very well to the ground state to ground state transition for even-even alpha emitters,<sup>8</sup> the known<sup>9</sup> alpha energies and partial half-lives were used in the Preston<sup>10</sup>

form of the formula to calculate the nuclear radii of a number of even-even nuclides in the range  $Z=84$  to 98. Using these radii, a value of  $r_0$  was calculated for each nuclide from the relationship  $\text{radius} = r_0 A^{1/3}$  and the plot in Fig. 1 of the average value of  $r_0$  ( $r_0$  generally decreasing with increasing  $Z$ ) for each element indicates a just discernible minimum or plateau at  $Z=96$ . The average value of  $r_0$  for each element was plotted because there is no discernible regular variation of  $r_0$  with  $A$  at constant  $Z$ .

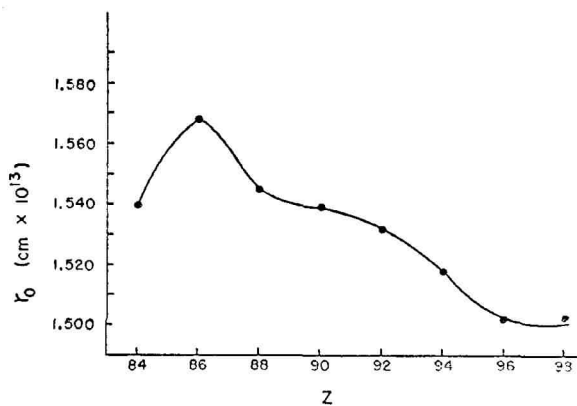


FIG. 1. Plot of average values of  $r_0$  vs  $Z$ . The following isotopes are included:  $\text{Po}^{212}$ ,  $\text{Po}^{214}$ ,  $\text{Po}^{216}$ ,  $\text{Po}^{218}$  for Po;  $\text{Em}^{220}$ ,  $\text{Em}^{222}$  for Em;  $\text{Ra}^{222}$ ,  $\text{Ra}^{224}$ ,  $\text{Ra}^{226}$  for Ra;  $\text{Th}^{224}$ ,  $\text{Th}^{226}$ ,  $\text{Th}^{228}$ ,  $\text{Th}^{230}$ ,  $\text{Th}^{232}$  for Th;  $\text{U}^{230}$ ,  $\text{U}^{232}$ ,  $\text{U}^{234}$ ,  $\text{U}^{235}$ ,  $\text{U}^{238}$  for U;  $\text{Pu}^{238}$ ,  $\text{Pu}^{239}$ ,  $\text{Pu}^{240}$ ,  $\text{Pu}^{242}$  for Pu;  $\text{Cm}^{242}$ ,  $\text{Cm}^{244}$  for Cm;  $\text{Cf}^{250}$ ,  $\text{Cf}^{252}$  for Cf.

The stable shell of 82 protons is attained upon completion of the  $h_{11/2}$  level and the spin of  $\text{Bi}^{209}$  (9/2) indicates that the 83rd proton begins the filling of the  $h_{9/2}$  level as might be expected. However, if the  $h_{9/2}$  level is raised in energy as more protons are added, so that the  $f_{7/2}$  and  $f_{5/2}$  are filled before the  $h_{9/2}$  levels, one might expect subshell effects at  $Z=90$  and 96. If the quantum states are filled in this order, the variation of  $r_0$  with  $Z$  should perhaps also indicate an effect  $Z=90$ . A careful consideration of the values of  $r_0$  in the region on both sides of  $Z=90$  points to a barely discernible plateau in the variation of  $r_0$  with  $Z$  at this atomic number.

In the case of  $Z=96$  there is an additional argument which points to the completion of a subshell here. The known odd-even isotopes of berkelium,  $\text{Bk}^{243}$  and  $\text{Bk}^{245}$ , are highly "hindered" in their most prominent modes of alpha decay, i.e., they decay much slower than the simple formula would indicate. This exceptional degree of hindrance is not observed for similar (odd-even) isotopes of any other odd  $Z$  alpha-particle emitter with the exception of bismuth ( $Z=83$ ) where the slowed rates of decay are presumably to be associated with the closed proton shell (and consequent shrunken radius) at  $Z=82$ .

There are other lines of evidence which may also point to the filling of the  $f_{7/2}$  and  $f_{5/2}$  before the  $h_{9/2}$  proton states. The spins of  $\text{Np}^{237}$ <sup>11</sup> and  $\text{Am}^{241}$ <sup>12</sup> are both 5/2 as expected on this basis. On the other hand, the spins of  $\text{Ac}^{227}$ <sup>13</sup> and  $\text{Pa}^{231}$ <sup>14</sup> have both been reported as 3/2, indicating perhaps that the odd proton occupies the  $p_{3/2}$  state, whereas spins of 7/2 and 5/2 corresponding to  $f_{7/2}$  and  $f_{5/2}$  states, respectively, would be expected. Whether or not this indicates a breakdown of the single-particle model, it does seem to indicate that arguments based on spin values cannot be conclusive here. It is interesting to add that a consideration of the systematics of beta radioactivity in this region also leads to the assignment of spectroscopic states in agreement with the suggested higher position of the  $h_{9/2}$  level energetically.

It is interesting to note that arguments based on spin values<sup>15</sup> indicate that in the case of neutrons the  $f_{7/2}$  level fills before the  $h_{9/2}$  level just after the completion of the major shell at 82 neutrons. Thus, the situation is analogous to that postulated for protons although the evidence is not clear on the relative position of the  $f_{5/2}$  and  $h_{9/2}$  neutron levels.