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ABSTRACT

The excitation functions for the (p,2n), (p,4n), (p,6n), and (p,8n) reactions on bismuth have been determined at high energy. A hypothesis is advanced to account for the ratio of the yields of the polonium products at high energies.

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INTRODUCTION

Relatively few investigations of excitation functions made at very high energies have been published, and in no case have the yields of a series of products of the (p,xn) reactions from a single target, such as the (p,n),(p,2n),(p,3n),(p,4n),(p,5n), etc., been reported for the hundred million volt range. As a result of the studies of the light isotopes of polonium,[1,2,3] it has become feasible to investigate such a series, produced by proton bombardment of Bi^{209}; thus with Bi(p,2n) (p,4n),(p,6n) and (p,8n) the products are observable through their distinctive alpha particles.

It was considered desirable to verify by another method the experimental work of Meinke, Wick and Seaborg[4] on the shape of the excitation curve, at high energy for the (p,xn) reactions.

II. EXPERIMENTAL

The excitation functions were determined by a series of bombardments at different bombarding energies, in the internal beam of the 184-inch Berkeley cyclotron. The results of each bombardment were calculated in terms cross-section by the expression $\sigma = n/NI$, where n is the number of atoms produced, N is the number of target atoms per cm^2, and I is the beam intensity in total atoms. These quantities were determined experimentally -- N by measuring the weight and area of the target, n by counting the polonium products and I by the use of a monitor which was bombarded simultaneously with the target. The monitor chosen was the 14.9 hr.

Na^{24} which is produced by the (p,3pn) reaction on Al^{27}. The excitation curve for Na^{24} has been determined by Stevenson and Folger,[5] so by using their cross sections and counting the Na^{24} activity, the total beam intensity could be calculated.

The targets were a thin layer of bismuth spread while molten on 5-mil aluminum foil for support. The targets were about 15-20 mg Bi/cm^2 in thickness, as determined by weighing the aluminum both before and after spreading the bismuth on it. In the bombardment, the 1-mil aluminum foil was clamped both in front and behind the bismuth target, and trimmed to cover the same area as the target. After bombardment, the 1-mil aluminum foil and the bismuth target were cut simultaneously from the target holder with a scapel. The strips of 1-mil aluminum foil were used to determine the beam intensity through the yield of 14.9 hr. Na^{24} produced during bombardment.

The aluminum foil with the bismuth coating was dissolved in 1-2 drops HNO_3 and 2 cc of concentrated HCl. The solution was evaporated to about 1 cc and the polonium was extracted with 20% tributyl phosphate in dibutyl ether.[2] Plates for counting were prepared by evaporation of the organic solvent on one-inch platinum counting disks. Yields of the various polonium isotopes were determined by pulse analysis of their alpha particles on a 48-channel differential pulse analyzer,[6] and comparison of the sample used for pulse analysis with a measured aliquot of the total polonium produced. Corrections were made in the yield of polonium for the geometric counting arrangement and decay from the end of bombardment. No corrections were necessary for decay during bombardment, since none of the bombardments was longer than 10 minutes.

Corrections were made in the yield of the Na^{24} monitor for geometry, absorption in air, window, covering material, and efficiency of the Geiger tube by counting a sample of RaE in equilibrium with RaF in the same Geiger counter and under the same conditions as the Na^{24} samples were counted. The absolute number of disintegrations of the RaE standard was determined by counting the RaF(Po^{210}) alpha particles from the same sample. The absolute number of disintegrations of the Na^{24} was calculated assuming the same ratio between the number of disintegrations and number of counts found in the RaE-RaF equilibrium mixture. There may be some error in this correction due to the difference in energies of the beta particle from RaE and Na^{24}(1.17 mev RaE, 1.39 mev Na^{24}) which would tend to overcorrect the yield of Na^{24}, and the absorption of the beta particles in the sample has been neglected. The determination of the beam is expected to be within ten percent of the true value.

III. RESULTS

The results obtained are given as apparent cross section, which is related to the true cross section by the equation:

$$\sigma_{\text{app}} = \sigma_{\text{true}} \; n_{\alpha}/\Sigma_{i} n_{i}$$

where $n_{\alpha}/\Sigma_{i} n_{i}$ is the fraction of atoms of the isotope of interest which decay by alpha emission. This is equivalent to plotting the yield of alpha emitters for the product under consideration, neglecting yield of the product which decays by electron capture paths. Since the branching ratios of the isotopes produced are unknown in most cases, it was decided to use this procedure.

The isotopes measured in this work are listed in Table I.

Table I

Isotope	Half-life	E_α(mev)	Mode of decay	Produced by
Po^{208}	2.9-yrs.	5.10	α (100%)	$Bi^{209}(p,2n)$
Po^{206}	9-day	5.21	α, electron capture	$Bi^{209}(p,4n)$
Po^{204}	3.8-hr.	5.37	α, E.C.	$Bi^{209}(p,6n)$
Po^{202}	50-min.	5.59	α, E.C.	$Bi^{209}(p,8n)$
Po^{200}	11-min.	5.84	α, E.C.	$Bi^{209}(p,10n)$

Table II gives the yields of the various isotopes and the energy of the bombarding protons. Where more than one determination has been made at a given energy, the value given is an average value.

The data of Table II are plotted in Figures 1 and 2 to give the excitation functions, Fig. 1 showing the (p,2n) and (p,4n) curves, while Fig. 2 shows the (p,6n) and (p,8n) curves. Fig. 3 is a plot of the (p,8n) reaction with the cross section on a linear scale. It may be seen that the shape observed for the excitation function for the (p,6n) reaction on thorium determined by Meinke, Wick, and Seaborg[4] persists for the (p,8n). The curve for the (p,8n) reaction may be

Table II

σ in millibarns

E(mev)	Po^{208}	Po^{206}	Po^{204}	Po^{202}
70	15	2.5	0.70	<0.0075
75	14	1.2	0.54	0.12
80	10	1.2	0.52	0.26
85	12	1.6	0.46	0.41
90	14	1.25	0.65	1.44
100	8.7	1.1	0.44	1.1
110	11.8	1.4	0.42	0.91
120	5.8	0.57	0.21	0.51
150	8.8	1.0	0.20	0.40
200	3.7	0.50	0.08	0.17
225	-	-	0.09	0.17
250	-	-	0.12	0.22
300	2.5	0.27	0.07	0.08
345	3.0	0.21	0.05	0.07

virtually superimposed on Meinke's curve for the (p,6n) by shifting the energy scale about 15 mev.

The accuracy of the apparent cross section determinations is not as high as had been hoped for, judged on the basis of reproducibility. However, the maximum deviation found between two identical measurements is 50 percent, and it is expected that most measurements will be uncertain by no more than 20 percent. The curves are drawn by weighing some points more heavily than others--it is usually apparent from a study of the (p,2n) or (p,4n) curves whether a particular determination is expected to be in error badly and in what direction the error is likely to be. The (p,2n) and (p,4n) excitation functions appear to be slowly varying functions at energies of this magnitude.

At energies well above the maximum yield of Po^{202}, it is found that the ratio of yields of the various isotopes to yields of Po^{208} at the same energy is essentially constant for each isotope. Table III shows the ratios of the apparent cross sections of Po^{206}, Po^{204}, and Po^{202} to the cross section of Po^{208}. The constancy of the ratios in each case is well within experimental error, although a possible deviation at full energy may be noted.

The constancy of the various ratios in Table III definitely shows that the yields of the (p,xn) reactions are in a constant ratio at bombarding energies past the peak yields and that the ratio of the yields is not energy dependent, even though any absolute yield is quite dependent on energy. One expects a regular variation in the cross sections, at a particular energy, as x changes. An hypothesis, attractive in its

Table III

E(mev)	σ_{app} Po^{206}/σPo^{208}	σ_{app}Po^{204}/σPo^{208}	σ_{app}Po^{202}/σPo^{208}
70	0.17		
75	0.13		
80	0.12		
85	0.14		
90	0.09		
100	0.12		
110	0.12		
120	0.10		
150	0.11	0.023	0.045
200	0.13	0.022	0.046
300	0.11	0.028	0.035
345	0.07	0.020	0.024
Average	0.12	0.023	0.038

simplicity, is that the cross section is independent of x, for a considerable range of values of x, as long as one is well above the maxima found at energies several mev above the respective thresholds. On this hypothesis, the ratios of the apparent cross section of each isotope to the cross section of Po^{208} represents the degree of alpha decay each isotope undergoes. On this basis, since the ratio of σapp for Po^{206} to σ for Po^{208} is 0.12, Po^{206} decays about 12% by alpha decay. It is interesting to note that Fung[7] has obtained the average value of 0.116 for the ratio of σapp for Po^{206} to σ for Po^{208} in a study of the (d,xn) excitation functions on bismuth.

The alpha branching ratios for Po^{206}, Po^{204}, and Po^{202} obtained by the hypothesis of equal yields are shown in Table IV, together with the partial alpha half-lives calculated using these branching ratios. A plot of the partial alpha half-lives vs. alpha-decay energy for the even-even polonium isotopes is shown in Fig. 4. The solid curve has been calculated[8] for even-even isotopes whose nuclear radius follows the relation $r = 1.48A^{1/3} \times 10^{-13}$ cm. It will be noted that all the isotopes whose branching ratios have been calculated on the basis of equal yields lie above the curve indicating, if these values are correct, that these isotopes show an abnormally small nuclear radii, but with deviations less than those at Po^{208} and Po^{210}.

Table IV

Isotope	Alpha-Disintegration Energy(mev)	α Branching %	Partial α-Half-life
206	5.32	12	82 days
204	5.48	2.3	180 hrs.
202	5.82	3.8	14 hrs.
200	5.96	~20	55 min.

No proof of the hypothesis used to obtain these branching ratios can be given, although a study of the (p,xn) reactions on thorium would yield a direct verification of its plausibility. The data plotted in Fig. 4 indicate that an error of a factor of 10 or more is very unlikely in these cases. It is hoped that further work will be undertaken to clear up this point.

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Note Added May 23, 1951

Since the bismuth foil is in contact with the aluminum monitors, the effect of multiple traversals by the particles of the beam is to cause an uncertainty in the energy spectrum of the incident particles. Except for edge effects, which are minimized by careful trimming, the beam through the bismuth in each case is the same as that through the monitors. Since the cross sections are slowly varying functions of energy in the energy region considered in this paper, the error due to energy spread of the beam is probably not important.

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[1] Templeton, Howland, and Perlman, Phys. Rev. 72, 758 (1947).

[2] D. G. Karraker and D. H. Templeton, Phys. Rev. 81, 510 (1951)

[3] Karraker, Ghiorso, and Templeton, Phys. Rev. (to be published).

[4] Meinke, Wick, and Seaborg, University of California Radiation Laboratory Report UCRL-868 (1950).

[5] P. C. Stevenson and R. L. Folger (unpublished).

[6] Ghiorso, Jaffey, Robinson, and Weissbourd, National Nuclear Energy Series, Plutonium Project Record, Vol. 14B, "The Transuranium Elements: Research Papers," Paper No. 16.8 (McGraw-Hill Book Company, Inc., New York, 1949).

[7] S. C. Fung, unpublished data.

[8] I. Perlman and T. J. Ypsilantis, Phys. Rev. 79, 30 (1950).

LIST OF ILLUSTRATIONS

Figure

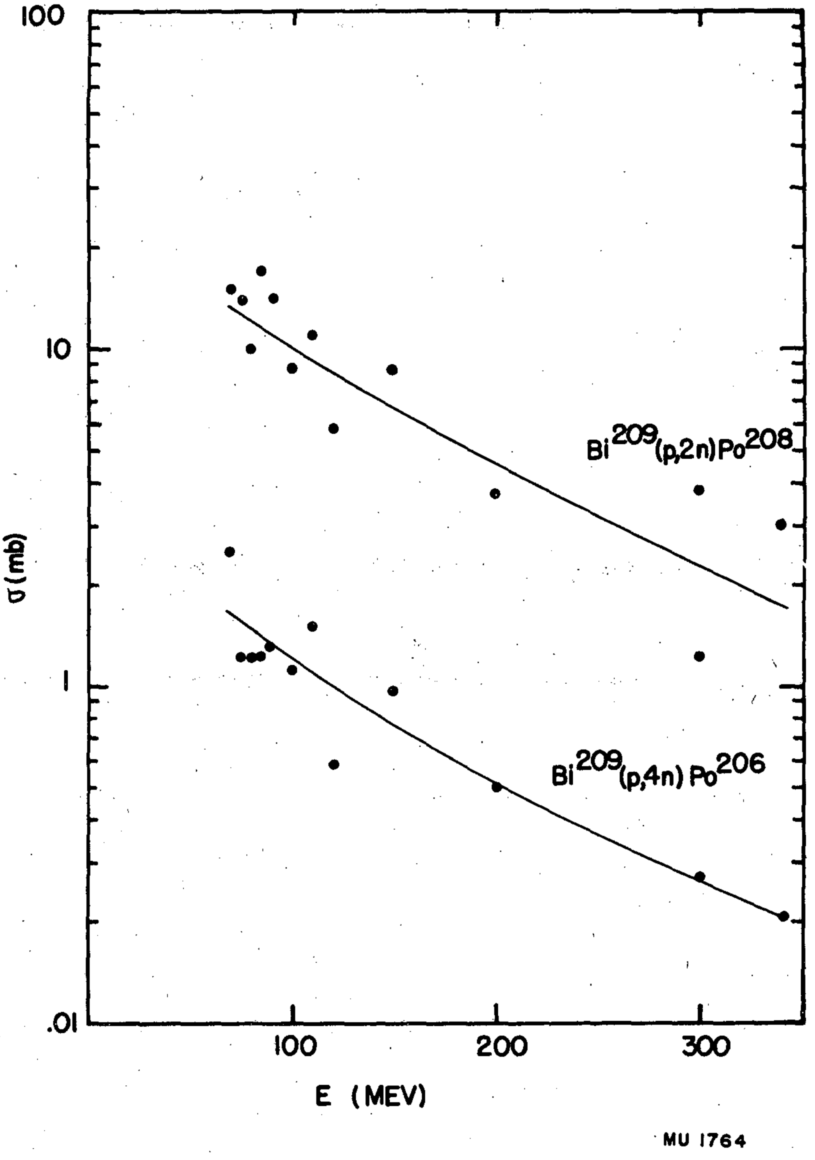

100
10
1
.01
σ (mb)
100
200
300
E (MEV)
$Bi^{209}(p,2n)Po^{208}$
$Bi^{209}(p,4n)Po^{206}$
MU 1764

Fig. 1

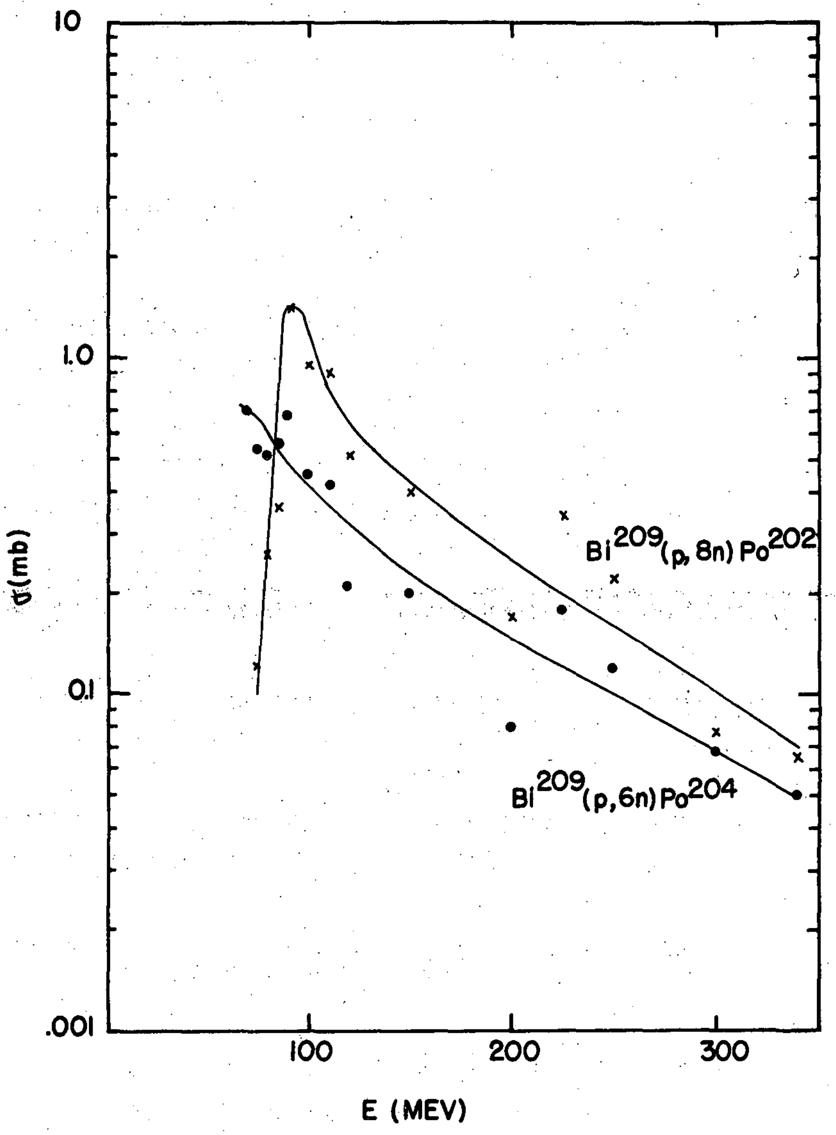

10
1.0
0.1
.001
σ (mb)
100
200
300
E (MEV)
$Bi^{209}(p,8n)Po^{202}$
$Bi^{209}(p,6n)Po^{204}$
MU 17.65

Fig. 2

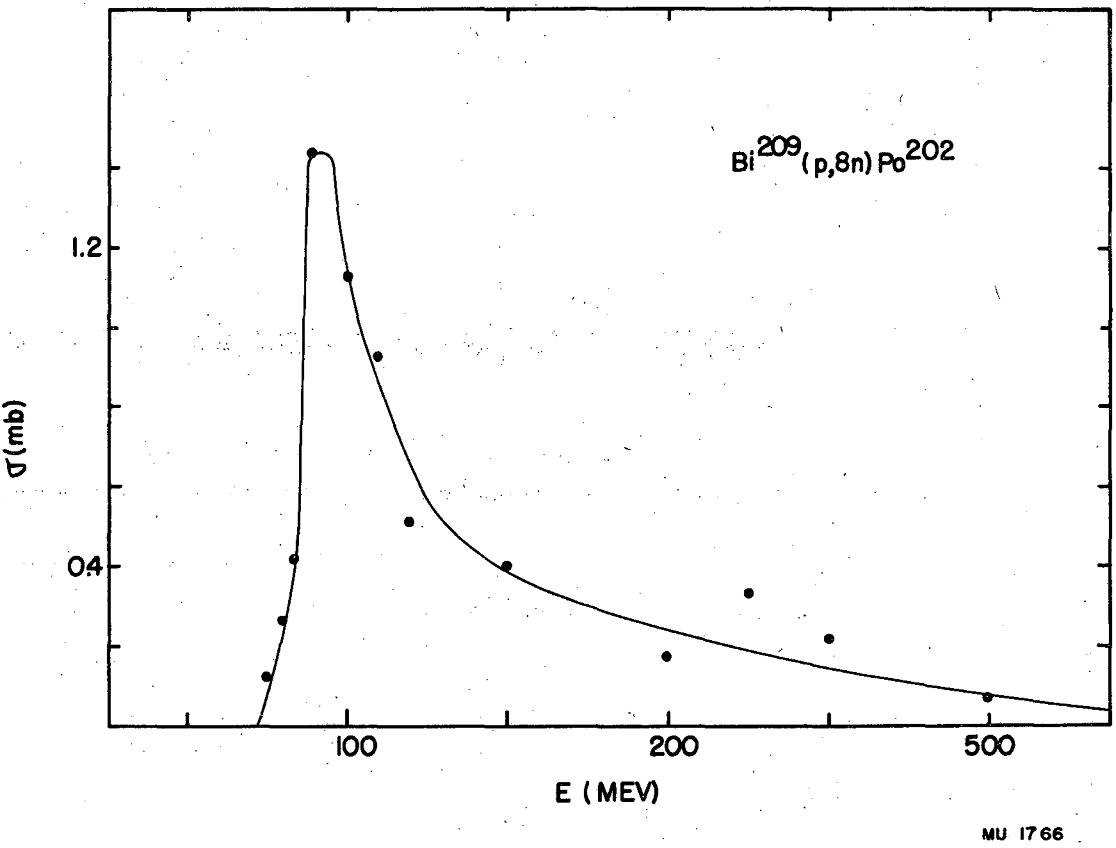
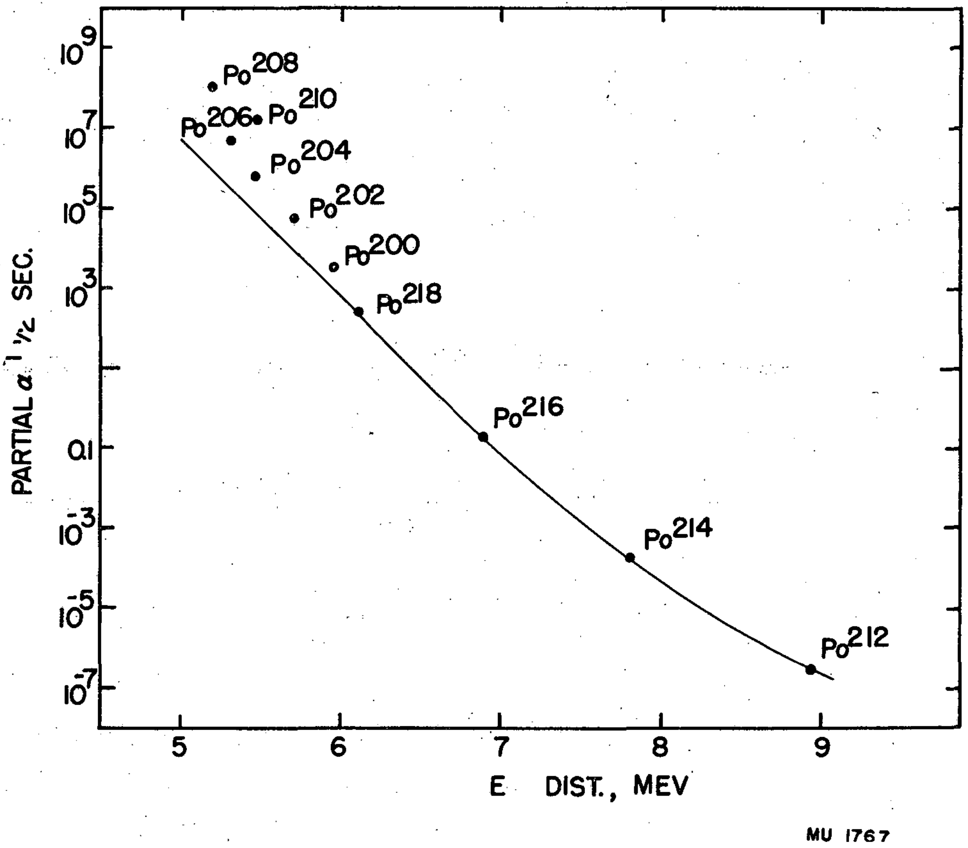


Fig. 3



PARTIAL α $T_{1/2}$ SEC.
10^9
10^7
10^5
10^3
0.1
10^{-3}
10^{-5}
10^{-7}
5
6
7
8
9
E DIST., MEV
Po208
Po206
Po210
Po204
Po202
Po200
Po218
Po216
Po214
Po212
MU 1767

Fig. 4