Spallation of Bismuth by 380-Mev Protons

E. T. HUNTER* AND J. M. MILLER
Chemistry Department, Columbia University, New York, New York
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Bismuth was bombarded by the 380-Mev proton beam of the Nevis Cyclotron to determine production cross sections of various spallation products. Products isolated and assayed were radioactive isotopes of polonium, bismuth, lead, thallium, mercury, gold, and platinum. Each of these elements was separated from the bismuth targets, carefully purified, and counted using a NaI(Tl) crystal scintillation counter having 4π geometry and $99_{-4}^{+1}\%$ efficiency for photons between 20 and 100 kev.

A mass-yield curve was constructed from these results and compared to the mass-yield curve constructed from available Monte Carlo calculations. The calculated data fit the experimental data quite well for products within about 20 mass number units of the target. A lower limit to the total spallation cross section of 1.14 ± 0.05 barns was obtained.

I. INTRODUCTION

PREVIOUS investigations[1–3] of the spallation of bismuth suffered from the lack of detailed information on the decay schemes of the product nuclides and the concomitant uncertainty in the counting efficiencies of the detectors that were employed. With the present availability of detailed information on more decay schemes and through the use of a 4π scintillation counter, it became possible to obtain a clearer picture of the spallation of the heavy element bismuth. The results of this investigation may then be compared with recent calculations[4–6] designed to simulate the two steps postulated for high-energy reactions: the intranuclear cascade generated by the incident proton followed by the evaporation of particles from the excited residual nucleus.

II. EXPERIMENTAL

A. Irradiations

Bismuth targets were irradiated for times varying from one to thirty minutes by placing them directly in the circulating beam of the Columbia University Nevis Synchrocyclotron at a radius corresponding to an incident proton energy of 380 Mev. Two types of targets were used: BiOCl powder wrapped in aluminum foil, and bismuth metal foil in a stack of aluminum foils. From the latter bombardments, the cross section for the production of an internal monitor, Bi^{205}, was determined with respect to the known cross section of 11 mb for the $Al^{27}(p,3pn)Na^{24}$ reaction.

B. Chemical Procedures

The target was dissolved in concentrated HNO_3 and divided into aliquots from which polonium, bismuth, lead, thallium, mercury, gold, and platinum fractions were isolated.

Polonium was extracted from a $6N$ HCl solution with an 80% dibutyl ether–20% tri-*n*-butyl phosphate mixture. Concentrated HNO_3 was used to return the polonium to an aqueous layer to which was added Bi^{+++} carrier. Bismuth phosphate was then precipitated, carrying some of the polonium which was counted in this form.

Bismuth purification consisted of the precipitation of $BiPO_4$ from $1N$ HNO_3, solution in HCl, and reduction with nickel powder. The bismuth-nickel mixture was then dissolved in concentrated HNO_3, and the above steps were repeated often enough to produce a radiochemically clean sample. Bismuth was counted as the phosphate.

The precipitation of $PbCrO_4$ and its subsequent conversion to $PbSO_4$ were the steps used to clean lead, which was finally counted as the chromate.

Thallium was oxidized with bromine water and nitric acid and extracted into diisopropyl ether. The ether was evaporated, the thallium reduced to Tl^+ with hydrazine hydrate and precipitated as TlI, in which form it was counted.

Mercury and gold were extracted together into ethyl acetate from the original nitric acid solution, to which was added a trace of chloride. Mercury was removed from the organic layer with $3N$ HCl, precipitated as HgS, and counted. The remainder of the organic layer was evaporated, the gold was taken up with aqua regia, then carefully reduced with hydrazine hydrate and counted as the metal.

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[1] W. E. Bennett, Phys. Rev. **94**, 997 (1954).

[2] Vinogradov, Alimarin, Baranov, Lavrukhina, Baranova, and Pavlotskaya, *Proceedings of the Conference of the Academy of Sciences of the U.S.S.R. on the Peaceful Uses of Atomic Energy, Moscow, July, 1955* (Akademiia Nauk, S.S.S.R., Moscow, 1955) [English translation by Consultants Bureau, New York: U. S. Atomic Energy Commission Report TR-2435, 1956].

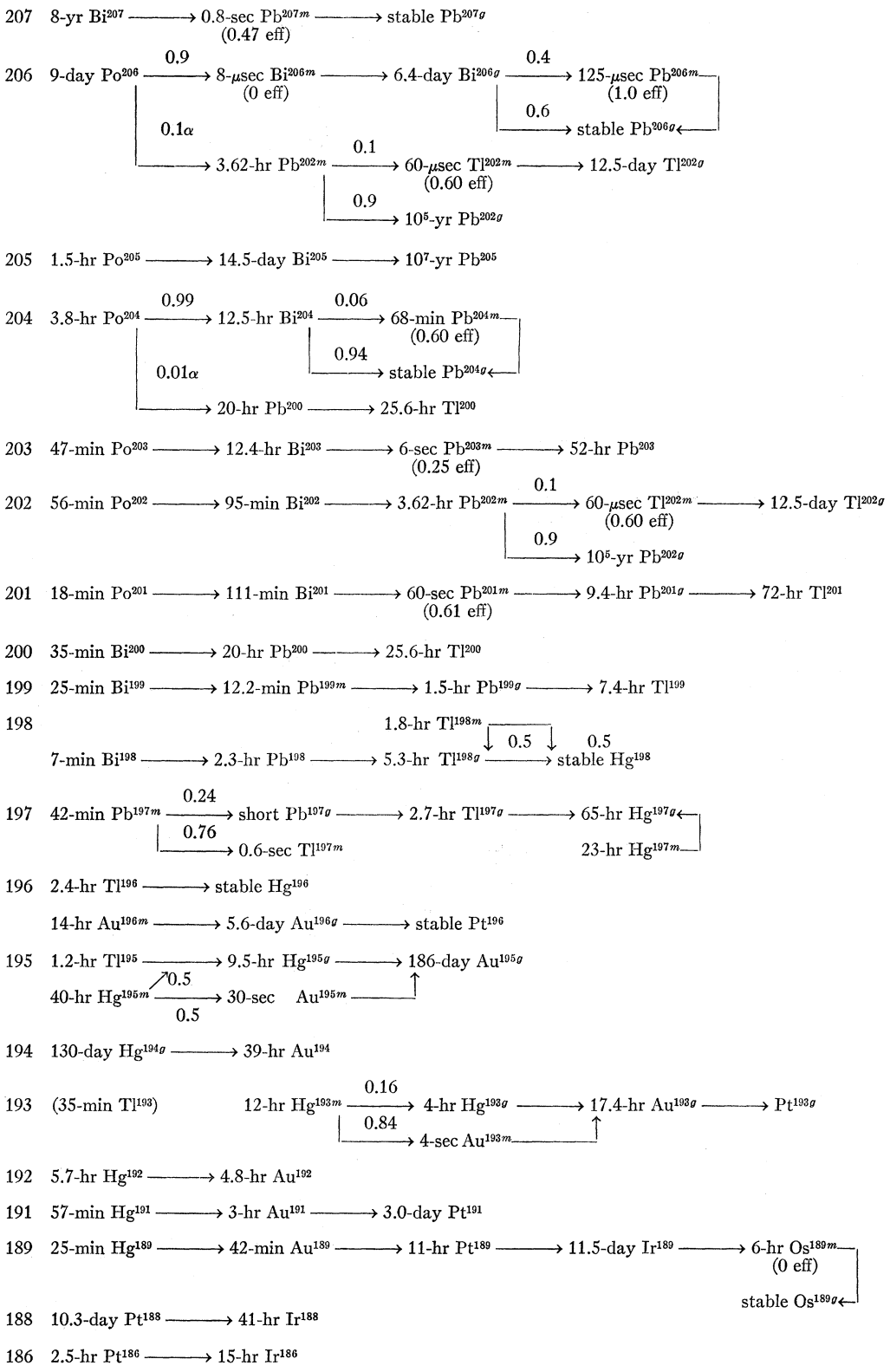
[3] Murin, Preobrazhensky, Yutlandov, and Yakimov, *Proceedings of the Conference of the Academy of Sciences of the U.S.S.R. on the Peaceful Uses of Atomic Energy, Moscow, July, 1955* (Akademiia Nauk, S.S.S.R., Moscow, 1955) [English translation by Consultants Bureau, New York: U. S. Atomic Energy Commission Report TR-2435, 1956].

[4] Metropolis, Bivins, Storm, Miller, Friedlander, and Turkevich, Phys. Rev. **110**, 185 (1958), and J. M. Miller (private communication).

[5] Dostrovsky, Rabinowitz, and Bivins, Phys. Rev. **111**, 1659 (1958).

[6] Dostrovsky, Fraenkel, and Rabinowitz, *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958* (United Nations, Geneva, to be published).

TABLE I. Decay schemes.



Gold and mercury were removed from the platinum fraction by extraction into ethyl acetate. The platinum was then reduced with $SnCl_2$ to $PtCl_4^=$, which was extracted into ethyl acetate. The organic layer was then evaporated to dryness, the platinum was taken up in aqua regia and precipitated as Rb_2PtCl_6, in which form it was counted.

The samples were prepared for counting by filtering onto retentive filter paper in a chimney arrangement which gave an evenly distributed sample of about $\frac{3}{8}$-in. diameter. This sample was then covered on both sides by small circles of cellophane, and Scotch brand tape was used to seal in the entire filter paper circle. This prevented the loss of any sample during the counting, and the use of cellophane allowed the sample to be easily recovered for chemical yield analysis. Colorimetric analytical procedures found in Sandell[7] were used in the chemical yield determinations.

C. Counting

Radionuclides that are expected to be produced in the spallation of bismuth are all on the neutron-deficient side of stability and, therefore, decay by electron capture. Each electron capture is often accompanied by a group of gamma rays, each with its own conversion coefficient, so that any counter other than one with a 100% counting efficiency for every electron capture will have a different efficiency for each one. This would lead to an insurmountable counting problem. Therefore, we have used a well-type double-crystal scintiallation counter, which was designed to have a 4π geometry, and which has a $99_{-4}^{+1}\%$ counting efficiency for a photon with energy between 20 and 100 kev.

D. Decay Curve Analysis

Serious difficulties arose in the analysis of the complex decay curves found in this work. These curves contained contributions from up to 10 independent decay chains of 1 to 4 members each. The several methods of attack employed were ordinary graphical resolution, least squares resolution over small parts of the total decay curve for 2 to 4 components, and an analytical method for curves in which only two activities of similar half-life were present. All of these methods, however, depend quite heavily on the assumed decay schemes, branching ratios, and half-lives. The decay schemes, half-lives, branching ratios, and counting efficiencies for the isomeric transitions without internal conversion that are listed in Table I are those used in this study. Most of the information comes from *Table of Isotopes*,[8] but many of the half-lives that were used are those found in this work.

[7] E. B. Sandell, *Colorimetric Determination of Traces of Metals* (Interscience Publishers, Inc., New York, 1950).

[8] Strominger, Hollander, and Seaborg, Revs. Modern Phys. **30**, 585 (1958).

TABLE II. Experimental and calculated cross sections.

Nuclide	Experimental cross section (in mb)	Number of determinations	Calculated cross section (in mb)
Po^{207}	0	3	12.3± 4.8
Bi^{207}	15.7 ± 3.6	9	15.3± 5.3
Po^{206}	7.7 ± 5.6	2	7.0± 3.6
Bi^{206}	49.3 ± 5.9	20	33.9± 7.9
Po^{205}	12.9 ± 9.4	2	16.8± 5.6
Bi^{205}	50.0 ± 7.0	11	21.6± 6.3
Po^{204}	8.9 ± 6.5	2	19.9± 6.1
Bi^{204}	37.1 ± 3.2	6	16.9± 5.6
Po^{203}	12.5 ± 9.1	2	13.3± 5.0
Bi^{203}	47.6 ± 7.6	6	25.0± 6.8
Pb^{203}	14.0 ± 2.7	4	16.2± 5.5
Po^{202}	5.2 ± 3.8	2	21.1± 6.3
Bi^{202}	55.8 ± 9.4	3	36.6± 8.2
Pb^{202m}	0	2	20.5± 6.2
Tl^{202}	4.42± 0.59	3	6.1± 3.4
Po^{201}	13.3 ± 9.7	2	16.0± 5.5
Bi^{201}	49.6 ± 4.4	3	27.5± 7.1
Pb^{201}	24.5 ± 6.6	3	19.0± 6.0
Tl^{201}	15.1 ± 3.4	3	9.0± 4.1
Hg^{193}	~7	1	8.7± 4.0
Au^{193}	~0.6 ± 0.6	3	0.7± 1.2
$Pt^{193m,191}$	~9	3	1.1± 1.4
Hg^{192} [a]	22.0	1	61.6±17.8
Au^{192}	14.2 ± 1.3	2	3.4± 2.5
Hg^{191} [a]	21.9	1	55.6±18.6
Au^{191}	17.0	1	10.5± 4.4
Hg^{190} [a]	39.8	1	51.2±17.3
Hg^{189} [a]	1.05	1	39.7±15.7
Au^{189}	7.8	1	11.7± 4.7
Pt^{189}	16.8	1	1.8± 1.8
Pt^{188} [a]	20.8 ± 5.9	3	47.6±19.6
Pt^{186} [a]	28.1	1	32.6±15.6
Po^{200}	10.0 ± 7.3	1	21.8± 6.4
Bi^{200}	64.4	1	30.2± 7.5
Pb^{200}	7.5 ± 5.0	4	21.1± 6.3
Tl^{200}	13.5 ± 0.8	3	7.3± 3.7
Bi^{199} [a]	68.6	1	38.9±11.8
Pb^{199}	13.7	1	30.1± 7.5
Tl^{199}	2.52± 1.37	2	12.6± 4.9
Bi^{198} [a]	60.1	1	34.2±11.0
Pb^{198}	26.9	1	35.6± 8.1
Tl^{198m}	25.8 ± 3.2	3	12.3± 4.8
Pb^{197m} [a]	12.5	1	62.6±17.8
Tl^{197}	~0	3	25.6± 6.9
Hg^{197}	4.65± 2.83	3	3.8± 2.7
Tl^{196} [a]	62.5 ±18.4	3	77.8±22.5
Au^{196}	0.49± 0.07	3	0.1± 0.3
Tl^{195} [a]	62.3 ±15.2	3	79.2±22.2
Hg^{195m}	3.89± 1.87	2	7.3± 3.7
Au^{195}	~0	3	0.8± 1.2
$Tl^{194,3,2}$ [a]	73.3 ±21.1	2	190 ±52
Hg^{194}	<0.5	3	7.7± 3.8
Au^{194}	1.30± 0.17	3	1.1± 1.4

[a] Cumulative cross section.

III. RESULTS

The cross sections for the production of various nuclides in the irradiation of bismuth with 380-Mev protons are listed in Table II. The errors quoted are root-mean-square errors based upon multiple determinations.

Several different cross sections were observed for the production of Bi^{205} and Po^{205} in samples with various separation times of Po from Bi. These observed cross sections were, therefore, equal to $\sigma_{Bi}+\sigma_{Po}[f(t,\tau)]$, where $f(t,\tau)$ is a known function of the time of irradi-

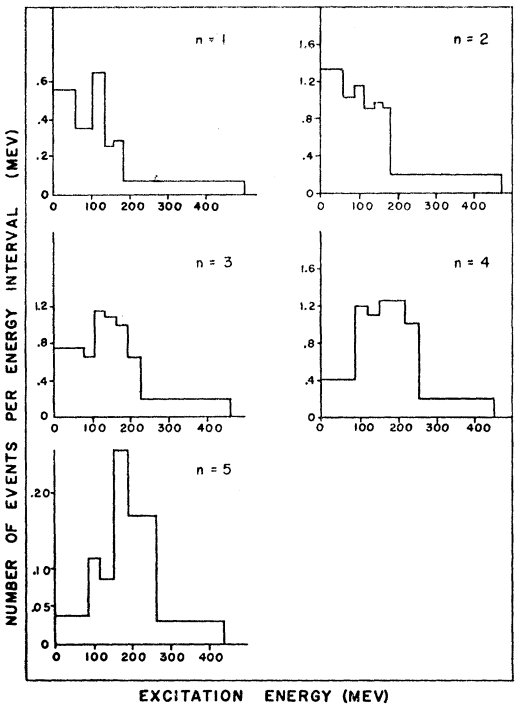


FIG. 1. Number of nuclei in various excitation energy intervals as a function of the number of nucleons, n, knocked out in the prompt cascade.

ation, τ, and the time elapsed between the end of irradiation and chemical separation, t. Solving these equations (one for each sample) by least squares for $\sigma_{Bi^{205}}$ and $\sigma_{Po^{205}}$ gave values of 50.0±7.0 and 12.9±9.4, respectively. The cross sections for the production of the other polonium isotopes were all determined relative to that of Po^{205}; therefore, their errors are at least that for Po^{205}.

A low yield of Tl^{197} has also been observed by Metzger[9] in another investigation and is possibly the result of an unobserved long-lived isomer. If it were the result of an unobserved short-lived isomer, the apparent cross section of Hg^{197} should be much higher than it is.

A 35-minute activity observed in the thallium fraction is probably a mixture of activities from $Tl^{194,193,\cdots}$ for which there are no data as to half-lives or decay schemes. The apparent cross section of 73.3±21.1 is therefore assigned to the production of this mixture.

Only the sum of the cross sections for the production of Pt^{191} and Pt^{193m} may be obtained since these nuclides have similar half-lives and could not be resolved.

Pb^{202} has a 3.62-hour isomeric state and a 10^5-year ground state. An unobservably small cross section is found for the isomeric state; because of the long half-life, nothing could be learned about Pb^{202g}. Therefore, for this nuclide and for several others with quite long half-lives no cross sections could be obtained.

[9] A. E. Metzger, Nevis Cyclotron Laboratory Report No. 66 (to be published).

IV. DISCUSSION

Spallation reactions in this energy range are considered to occur in two stages by the mechanism proposed by Serber[10]: a prompt intranuclear cascade initiated by the bombarding particle in a knock-on type process followed by a much slower de-excitation of the residual nucleus by the evaporation process. The results of this experiment may be compared with the results of Monte Carlo calculations designed to simulate the two independent steps of this mechanism.

About 1000 intranuclear cascades representing the irradiation of Bi^{209} with 450-Mev protons have been followed by the Monte Carlo method.[4] The mass, charge, and excitation energy of each residual nucleus were tabulated. Figure 1 shows histograms of the number of nuclei in various excitation energy intervals as a function of the number of nucleons knocked out in the prompt cascade.

The subsequent de-excitation of each of these residual nuclei either by evaporation or by evaporation and fission must then be considered. Both of these processes have also been studied by the Monte Carlo method. Evaporation calculations have been made[5] which give the average number of neutrons, protons, alpha particles, deuterons, and tritons that are evaporated from Ta^{181} with various initial excitation energies. These evaporation results for stable Ta^{181} are used as an approximation to the actual situation in which the residual nuclei are probably not all stable. In the total mass-yield curve which will be constructed from these calculated results, this difference in the nuclei prior to evaporation will have little effect because the mass lost by evaporation is rather insensitive to the isotopic number of the initial excited nucleus. However, as the starting nucleus becomes further removed from stability, the relative probabilities of losing neutrons and protons are altered considerably, and, therefore, the further that starting nuclei are removed from stability, the less reliable will be the calculated isotopic yields shown with the results in Table II. By use of the available data for the evaporation of particles from excited Ta^{181}, the product nucleus that results from the evaporation of particles from each excited nucleus remaining after the prompt cascade was found and was tabulated by A and Z. These results must then be corrected for the fission-evaporation competition. Monte Carlo calculations for this fission-evaporation competition have been made[6] and from them it is possible to obtain a rough correction for each of the evaporation chains. The final mass-yield curve obtained from these calculations is shown in Fig. 2 along with the observed experimental results. The fact that the Monte Carlo data refer to 450-Mev protons rather than 380-Mev incident protons is probably the main cause of the deviation between the experimental and calculated curves at ΔA greater than about 20. But in the

[10] R. Serber, Phys. Rev. **72**, 1114 (1947).

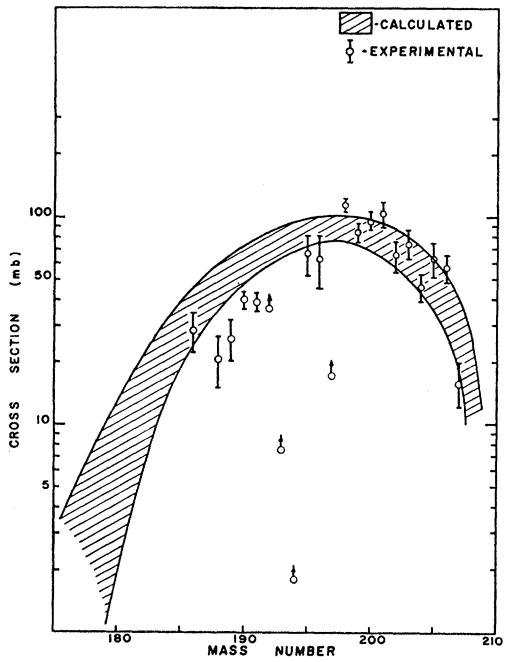


FIG. 2. Mass-yield curve from Monte Carlo calculations compared with experimental results.

main, the Monte Carlo calculation of the mass-yield curve fits the data quite well. However, the agreement between experiment and theory for individual product cross sections is not as satisfactory reflecting their greater sensitivity to the assumptions that were made.

The formation of stable nuclei in the spallation of bismuth is not probable because of the large neutron to proton evaporation ratio from the excited nuclei remaining after the fast cascade[5]; however, the lighter stable isotopes of elements such as mercury and platinum may be formed to some extent. As a result of this formation of stable nuclides, of nuclides having very long half-lives whose cross sections therefore cannot be determined, and of elements with Z less than 78, which were not investigated in this work, the total observed cross section is a lower limit to the true spallation cross section. This experimentally observed value is 1.14 ± 0.05 barns. This is to be compared with a geometric cross section of 1.86 barns ($r_0=1.3\times10^{-13}$ cm), a Monte Carlo value of 1.58 barns (total inelastic cross section minus fission cross section), and a value of 1.35 barns reported by Steiner and Jungerman[11] for the spallation cross section of bismuth with 350-Mev protons.

These results are also of interest in connection with the analysis of the high-energy fission process.[6] From this analysis it is found that several nuclei are the predominant fissioning species in the high-energy fission of bismuth. Therefore, the spallation cross sections for the production of nuclides near these should be of the same order as their contribution to the fission cross section. These fissioning nuclei have cross sections for fission of 5 mb or less which is consistent with the general pattern of spallation cross sections.

V. ACKNOWLEDGMENTS

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[11] H. M. Steiner and J. A. Jungerman, Phys. Rev. **101**, 807 (1956).