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PRODUCTION OF At^{209} *AND* At^{207} *BY HIGH-ENERGY PROTON BOMBARDMENT OF* Bi *AND* Pb

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The yields of At^{209} and At^{207} relative to the yield of At^{211} were determined when Bi and Pb were bombarded with 120—660-MeV protons. The astatine was separated chemically, and the relative yields were established from the α spectra measured with a gridded ionization chamber. The At^{209}/At^{211} yield ratio from bismuth was 0.64 ± 0.06 and 0.72 ± 0.06 for 120- and 660-MeV protons, respectively; the corresponding At^{207}/At^{211} yield ratios were 0.30 ± 0.03 and 0.51 ± 0.04. Bombardment of lead with 200- and 660-MeV protons and with 400-MeV deuterons resulted in the At^{209}/At^{211} yield ratios 1.31 ± 0.28, 1.43 ± 0.43, and 1.52 ± 0.25; for At^{207}/At^{211}, correspondingly, 0.28 ± 0.07, 0.61 ± 0.13, and 0.52 ± 0.15. The formation of these astatine isotopes is considered to result from secondary capture of "super-barrier" He^3, He^4, and Li nuclei produced in high-energy multinucleon interactions.

INTRODUCTION

THIS work continues a series of radiochemical investigations of the so-called secondary capture of "super-barrier" fragments (having energies above the Coulomb barrier). In the present case astatine (Z = 85) was produced by bombarding bismuth (Z = 83) and lead (Z = 82) with high-energy protons.

In earlier similar work[1-3] the production of the long-lived isotopes At^{211} and At^{210} was investigated. However, yields were determined with insufficient accuracy by analyzing the total α-decay curves to derive separate half-lives. Nevertheless, the analysis of the secondary-reaction cross sections,[1-4] evidently $(\alpha, 2n)$ and $(\alpha, 3n)$ in the case of bismuth and a set of reactions from $(Li^6, 3n)$ to $(Li^7, 5n)$ in the case of lead, yielded information regarding the energy spectrum of the captured fragments in the interval from 20—30 to 50 MeV.

The present work was undertaken to determine the yields of the relatively lighter secondary-reaction products At^{207} and At^{209}, which are formed through the capture of fragments having higher energies ($\geq$ 40 MeV). We determined the yields by measuring the α spectra, completely excluding errors involved in determining chemical yields and in monitoring the bombarding beam.

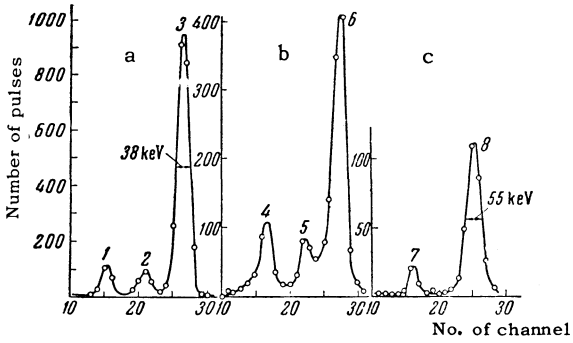
EXPERIMENTAL PROCEDURE

We used samples of high-purity bismuth,[2] obtained from the State Rare Metal Research Institute (Giredmet), and specially purified lead.[3] Purity checks showed that the samples of both metals contained practically no undesirable heavier elements. The samples were bombarded at the synchrocyclotron of the Joint Institute for Nuclear Research under the conditions described in [2,3]. Astatine was separated from the irradiated samples using a tellurium carrier, from which the astatine was extracted in diisopropyl ether and then reextracted.[5] Thin sources suitable for α-

spectrometer measurements were obtained by depositing astatine from the reextracted HCl solution on a polished silver surface.[6] The metallic silver disk in contact with the HCl solution was rotated at 60 rpm. Rotation during 20 min in a 1—2 N solution at 70°C resulted in more than 90% deposition of the astatine. The total time required to prepare a source was 2—3 hr, and usually 0.2—0.5 of the total amount of astatine in the irradiated sample was deposited on the disk.

The spectra were measured with a gridded ionization α-spectrometer[7] at different times; in this way both the energy and the half-life of each α group were determined. In the measurements performed on highly radioactive samples special mechanical collimators were used, resulting in considerable correction of the recorded spectra. These targets were also suitable for observing long-lived polonium daughter α emitters accumulated through K capture in some astatine isotopes. The weak sources were measured without collimators, thus somewhat impairing the accuracy of the results because of increased errors in analyzing the α spectra. No accumulation of long-lived polonium isotopes was observed in the weak samples.

The accompanying figure contains examples of the α spectra of astatine and polonium samples and their identification: a) with a collimator, b) without a collimator, and c) the α spectrum of long-lived polonium daughter isotopes. For 5.86-, 5.75-, and 5.64-MeV α particles we obtained the half-lives 7.1—7.6, 1.7—1.9, and 5.3—5.7 hr, respectively. The production of At^{211}, At^{207}, and At^{209} was thus established unambiguously.[8] The production of At^{211} is also confirmed by the presence of a 7.44-MeV α group from short-lived Po^{211} resulting from K capture in a fraction of the At^{211} nuclei. Long-lived daughter activities (Fig. 1c) comprised α peaks of Po^{210} (E = 5.3 MeV, T = 138 days) and Po^{208} (E = 5.1 MeV, T = 2.9 years).



α spectra of astatine and polonium and their identification. 1 – At^{209}, E_α = 5.64 MeV; 2 – At^{207}, E_α = 5.75 MeV; 3 – At^{211}, E_α = 5.86 MeV; 4 – AT^{209}, E_α = 5.64 MeV; 5 – At^{207}, E_α = 5.75 MeV; 6 – At^{211}, E_α = 5.86 MeV; 7 – Po^{208}, E_α = 5.1 MeV; 8 – Po^{210}, E_α = 5.3 MeV.

The relative yields of the different isotopes were computed from the contributions of different α groups using the customary radiochemical procedures, i.e., by measuring the accumulation in the course of irration and the subsequent decay and by considering the abundances of α decay (given as 41, ~5, and ~10% for At^{211}, At^{209}, and At^{207}, respectively, in [8]). It was assumed that At^{210} and At^{208} decay predominantly via K capture and that the daughters Po^{210} and Po^{208} decay only via α emission. The α count was corrected for the spectrometer dead time and for the decay of long-lived emitters during prolonged exposures.

RESULTS

The accompanying table gives the relative yields and possible deviations averaged over all runs. The relative yields were determined quite reliably for the Bi samples, most of which were measured using collimators. The individual measurements are in agreement within experimental error even in the cases when collimators were not used.

We used the relative yield $Y(At^{210})/Y(At^{211})$ as a control. At 660 MeV this ratio agrees well with the result obtained by other investigators at the same proton energy.[2] Our result at 120 MeV, however, differs somewhat from the result given in [2] for 130-MeV protons. Our result for $Y(At^{210})/Y(At^{211})$ at 120 MeV agrees well with that obtained by a French group[4] at 150 MeV.

The table shows that the At^{209}/At^{211} yield ratio from bismuth is nearly identical at 660 and 120 MeV, agreeing with the results in [4]. $Y(At^{207})/Y(At^{211})$ at 660 MeV is about 30% smaller, and at 120 MeV it is almost 50% smaller, than $Y(At^{209})/Y(At^{211})$. Our value for $Y(At^{207})/Y(At^{211})$ at 120 MeV is three times as large as the result given in [4]. Approximately the same

Sample	Particle and energy, MeV	Yield relative to At^{211}			
		At^{210}	At^{209}	At^{208}	At^{207}
Bi	p, 660	0.81±0.08	0.72±0.06	0.40±0.04	0.51±0.04
	p, 660 [2]	0.82±0.12	—	—	—
	p, 120	0.96	0.64±0.06	~0.5	0.30±0.03
	p, 130 [2]	0.63±0.10	—	—	—
	p, 150 [4]	1.02±0.20	0.81±0.22	0.22±0.05	0.10±0.04
Pb	p, 660	—	1.43±0.43	—	0.61±0.13 (0.62±0.12)
	p, 200	—	1.31±0.28	—	0.28±0.07 (0.56±0.25)
	d, 400	—	1.52±0.25	—	0.52±0.15 (0.72±0.10)
	α, 800	—	—	—	(0.71±0.35)

discrepancy between our results and those in [4] is observed for $Y(At^{208})/Y(At^{211})$.[1] The cause of these discrepancies is not clear. In connection with $Y(At^{207})/Y(At^{211})$ we may doubt the correctness of the results given by the French investigators, who obtained α peaks having a large half-width (~ 90 keV); however, their smaller result for $Y(At^{208})/Y(At^{211})$ can hardly be accounted for in the same way.

We see that the greatest yields of the heavier isotopes At^{211}, At^{210}, and At^{209} were obtained from Bi at both bombarding energies. The yield was reduced irregularly with decreasing isotope mass number. A reduction of the proton energy to one-sixth evidently does not affect the heavy-isotope yield ratio and reduces the relative yields of lighter nuclei to about one-half.

The values given for lead samples are less accurate because of the smaller statistics and the possibility of subjective errors in analyzing the spectra into groups. However, no large discrepancies between the individual measurements are observed, the deviations from the mean being less than ±30%. The yield of At^{209} is 30—50% greater than that of At^{211} and is almost independent of the proton energy. The probability of At^{207} production, which was determined more accurately in our work than in [3], is about 60% of the probability of At^{211} production at 660 MeV and is reduced to half of this ratio at 200 MeV.

As a check we repeated the determination of $Y(At^{207})/Y(At^{211})$ from lead samples by analyzing the decay curves, using an electronic computer and the method of least squares,[2] based on known α-decay constants;[8] the results are given in parentheses in the table. The lower accuracy of this ratio for 200-MeV protons and α particles results from the larger statistical errors involved in measuring weak activities in these cases.

The relative yields of At^{207} from lead which were calculated by the two different methods for the same bombarding energy are thus seen to agree within error limits. The constancy of $Y(At^{207})/Y(At^{211})$ for 660-MeV protons, deuterons, and high-energy α particles shows that the nature of the bombarding particles does not determine the yield ratio of these isotopes, and that probably only the energy of these particles is important.

[1]In calculating the At^{208} yield its half-life was taken, as in [4], to be 6.3 hr, because the admixture of the corresponding isomer (E = 5.65 MeV, T = 1.7 hr), if at all present, was insignificant, since it was not detected in our measurements.

[2]We wish to thank N. N. Govorun, E. A. Loginova, and I. N. Silin for analyzing the decay curves on the computer.

DISCUSSION OF EXPERIMENTAL RESULTS

When Bi is bombarded with high-energy protons astatine isotopes having mass numbers from 207 to 211 are most probably formed through the secondary capture of He^3 or He^4 fragments. The literature contains no information regarding the probabilities that He^3 and He^4 are emitted from Bi with high kinetic energy. Data can be found only for the relative yields of these nuclei with energies below the Coulomb barrier, and only for gold and uranium.[9] Here the He^3 abundance is estimated at not more than 10% of the number of α particles. Taking this estimate as correct for our case, we shall assume that when Bi is bombarded secondary (α, xn) reactions occur with x = 2, 3, 4, 5, 6, and shall neglect secondary reactions with He^3.

In the case of Pb there exists a wide range of secondary Li capture reactions, with the principal contributions evidently coming from the reactions (Li^6, n) to $(Li^7, 8n)$. For both Bi and Pb we thus encounter secondary reactions liberating different numbers of neutrons. These events are therefore captures of fragments having a fairly broad range of kinetic energies; for example, At^{207} can be formed only when α particles with 50—80 MeV are captured. The kinetic energy of a fragment is frequently several times greater than the total binding energy of its constituent nucleons. A satisfactory theoretical explanation has still not been found for the emission of fragments having such high energies. The principal difficulty arises in attempting to account for the fact that an incoming proton can transfer to a nucleonic subgroup several times the total binding energy of the latter without producing breakup of the subgroup. We attribute these events to direct multi-particle interactions between the incident proton and nucleons of the target nucleus. The possibility of multi-particle interactions is predicted by the meson theory of nuclear forces.[3] In our series of investigations of secondary interactions it was our principal aim to accumulate experimental data regarding this interesting phenomenon, and specifically to employ a single method (such as radiochemistry) in accumulating information on the probabilities of multi-particle interactions between protons having given energies and different numbers of intranuclear nucleons. Other modes of investigation are well supplemented by the study of secondary reactions, which often represents the only source of useful information.

It can be asked whether secondary reactions alone produce the observed astatine isotopes. It was noted in [1] that light astatine isotopes can

also be produced in bismuth by the capture of a high-energy proton with the emission of a π^- meson and several neutrons. A certain amount of At^{207} is probably formed in this way at 660 MeV, since its relative yield is 20% greater than that of At^{208}, and in this case the π^- meson is accompanied by three neutrons. In all other cases the given astatine isotopes are evidently formed only through secondary capture of "super-barrier" He^3, He^4, and Li. Since the At^{211} yield from Pb is approximately 1/15 the yield from Bi,[2,3] lithium capture accompanied by proton emission makes a negligibly small contribution to the production of astatine from bismuth.

We have calculated roughly the energy spectra of captured fragments from both Bi and Pb. We make no claim of precision, and have aimed only to obtain a provisional picture of the fragment spectrum in the range from 20 to ~ 100 MeV. The calculations were facilitated by a number of simplifying assumptions. Specifically, the fission of compound nuclei was not taken into account; as already noted, in the case of bismuth we neglected the contribution of He^3 capture. The energy spectrum was calculated from the schemes given in [1,4]. The calculations show that the relative yields of astatine isotopes from 120-MeV proton bombardment can be obtained by assuming an α-particle spectrum of the form $1/E^3$. The difficulties involved in calculating Γ_f/Γ_n reliably for interactions of Bi with α particles, especially at 60—90 MeV, resulted from the arbitrary selection of parameters.[10] The fact that the calculated excitation functions cannot be check experimentally prevents us from drawing more reliable conclusions regarding the spectrum of captured α particles. At the present time we can only affirm that the true α-particle spectrum must be harder than $1/E^3$, despite the fact that we omitted reactions with He^3, which should have the opposite effect on the spectrum.

In the case of lead the best agreement with experiment at 660 MeV is found in the Li spectrum given by evaporation theory for the parameters V = 6 MeV and τ = 9.5—11.5 MeV. Here $Y(At^{207})/Y(At^{211})$ (see Table II in [3]) agrees with our result, and $Y(At^{210})/Y(At^{211})$ is close to the experimental ratios $Y(At^{209})/Y(At^{211})$ and $Y(At^{210})/Y(At^{211})$.

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