INVESTIGATION OF SECONDARY (α, xn) REACTIONS IN BISMUTH

WANG YUNG-YÜ, V. V. KUZNETSOV, M. Ya. KUZNETSOVA, and V. A. KHALKIN

Joint Institute for Nuclear Research

Submitted to JETP editor February 5, 1960

J. Exptl. Theoret. Phys. (U.S.S.R.) **39**, 230-234 (August, 1960)

The absolute production cross sections and relative yields of At^{210} and At^{211} from bismuth irradiated by 120 — 660 Mev protons were determined by radiochemical means. In the given case the astatine isotopes are produced in the (α, xn) reaction when an α particle with an energy $E_\alpha > 20$ Mev, produced as a result of profound disintegration, is captured by a target nucleus. The cross sections for the production of α particles with energies $E_\alpha > 20$ Mev were calculated on the basis of the experimental data, and the shape of their energy spectrum was estimated.

INTRODUCTION

THE disintegration of complex nuclei by high-energy protons produces alpha particles, some of which possess a sufficiently high energy to give rise to secondary reactions in the target nuclei.[1-4] Determination of secondary-reaction yields enables one to reach conclusions about the shape of the spectrum of fast α particles, and to estimate the cross section for their production. In particular, this method was employed to determine the shape of the spectrum and the production cross section of fast α particles produced in the decay of bismuth[1] and gold.[2]

Despite the fact that in both references single isotopes and elements with near atomic numbers were studied, the shapes of the spectra of the fast alpha particles differ from each other considerably. In addition, the cross section for the production of particles with $E_\alpha > 20$ Mev in bismuth turned out to be 5 to 6.3×10^{-25} cm^2, which is more by a factor of 3 — 4 than the cross section obtained from the data of Perfilov and Ostroumov[5] who used the method of three photoemulsion layers.

In view of these differences, we thought it useful to repeat the work of Kurchatov et al.[1] and to clarify the reason for the observed discrepancies.

EXPERIMENTAL PART

We conducted our work with bismuth of high purity, the total impurity content of which did not exceed 10^{-4}%. The targets were irradiated in the synchrocyclotron of the Laboratory for Nuclear Problems at the Joint Institute for Nuclear Research. The proton energy was varied between 120 and 660 Mev by placing the target at different orbit radii of the accelerated protons inside the vacuum chamber of the accelerator.

To prevent losses of astatine due to heating of the target during irradiation, the bismuth was sealed in a quartz ampule with an outer diameter of (3.9 ± 0.1) mm, a wall thickness of 0.5 — 0.6, and a length of 30 mm. The metal occupied approximately half the volume of the ampule. The time of irradiation varied from 5 to 15 min. The proton-beam intensity was estimated from the production of Na^{24} in aluminum foil which was wrapped about the lower half of the ampule occupied by the metal.

Three hours after the irradiation, the astatine was extracted from the bismuth and precipitated together with elementary tellurium.[6] A suspension of elementary tellurium in water was uniformly deposited on a stainless-steel disc with a diameter of 2 cm and was dried at a temperature of 80 — 90°C. To prevent mechanical losses of tellurium during the measurements, the preparation was covered with a film of nitrocellulose (0.02 mg/cm^2).

The absorption of α particles in the tellurium layer and the covering film was determined experimentally. For this purpose we compared the α activity of equal amounts of astatine precipitated together with tellurium and covered with a film, and astatine deposited from a nitric-acid solution on a polished platinum disc. In the latter case the astatine preparation was without a carrier and there was no absorption of α particles in the layer. The experiment showed that the tellurium layer and the film which covered it absorbed 25%

of the α particles from At^{211} (E_α = 5.86 Mev) and Po^{211} (7.44 Mev), and 30% of the α particles from Po^{210} (5.3 Mev).

The alpha activity of tellurium preparations which contained astatine was measured with the aid of a scintillation counter consisting of an FÉU-19 photomultiplier with a ZnS(Ag) scintillator. The natural background of the measuring apparatus amounted to 10 — 20 pulses/sec. The total efficiency determined with the aid of an α standard (Pu^{239}, E_α = 5.15 Mev) was found to be ~35%. It was observed in the measurements that the activity of all preparations decreased with two half-lives: (7.3 ± 0.2) hr and 140 days; these are the half-lives of At^{211} and Po^{210}, respectively. Po^{210} is formed in the decay of At^{210} ($T_{1/2}$ = 8.3 hrs; K capture).

The activity of the monitor specimens (Na^{24}) was measured with a end-window MST-40 Geiger-Mueller counter. The total efficiency, determined with the $Sr^{89}\beta$ standard ($T_{1/2}$ = 51 days, E_α = 1.46 Mev), amounted to ~ 20%.

RESULTS

The absolute cross sections for the production of At^{210} and At^{211} for various energies of the bombarding protons E_p are listed in the table. The

E_p, Mev	σ (At^{211}), 10^{-29} cm^2	σ (At^{210}), 10^{-29} cm^2	$\sigma(At^{210})/\sigma(At^{211})$
130	0.52	0.33	0.63
170*	1.28	1.05	0.8
300	1.96	1.18	0.60
400	1.92	1.58	0.82
480*	2.52	2.10	0.8
530	2.82	2.28	0.81
580	2.26	1.67	0.66
660	2.60	2.14	0.82

*Corrected results of reference 1.

absolute cross section for the production of astatine isotopes from bismuth under the action of fast protons is here understood to be*

$$\sigma_{At}^{p} = n \int_{20\ Mev}^{\infty} \sigma_\alpha dE \int_{0}^{\infty} \frac{\sigma_{At}^{\alpha}}{-dE/dx} dE,$$

where n is the number of bismuth nuclei per cubic centimeter, σ_α is the cross section for the production of α particles on the target nuclei under the action of protons, σ_{At}^{α} is the cross section for the production of astatine isotopes under the action of α particles, and dE/dx is the energy loss of the α particles in the target material.

*Starting from the alpha spectrum, one can show that in the irradiation of thick specimens ($\delta > 1$ mm) σ_{At}^{p} is independent of the target thickness.

The ratio of the production cross sections of At^{210} and At^{211} is listed in the last column of the table. The production cross sections of the individual astatine isotopes are determined within a precision of ±30%, the error in the determination of their ratio does not exceed ±15%.

As can be seen from the results listed in the table, the production cross section of astatine isotopes increases by about a factor of five when the energy of the bombarding protons E_p is increased from 120 to 600 Mev. The cross section increases most rapidly up to energies of 300 — 400 Mev. The ratio of the cross sections $\sigma_{At^{210}}/\sigma_{At^{211}}$ as a function of E_p remains constant within the limits of the experimental error. This indicates that the shape of the spectrum of the α particles produced in the disintegration of bismuth remains practically unchanged in the 120 — 600 Mev range of proton energies.

The larger values of the production cross sections of At^{211}, and consequently also of the cross sections σ_α ($E_\alpha > 20$ Mev), obtained in reference 1, are explained by the fact that in the processing of the experimental data no allowance was made for the registration of At^{211} from the α activity of the daughter element Po^{211} ($T_{1/2}$ = 0.52 sec, E_α = 7.44 Mev). If the results of this work are suitably corrected, the obtained production cross sections of At^{211} are in good agreement with our data (cf. the table).

Comparison of our data on the cross sections of the secondary reaction products for E_p = 400 Mev with those of reference 2 for gold (E_p = 380 Mev) shows that the production cross sections of products of the secondary (α, 2n) reaction are in satisfactory agreement with each other; 1.92×10^{-29} cm^2 for bismuth, and $(2.25 \pm 0.5) \times 10^{-29}$ cm^2 for gold. The cross sections of the products of the (α, 3n) reactions differ, however, markedly, amounting to 1.58×10^{-29} cm^2 for bismuth and $(4.2 \pm 0.8) \times 10^{-29}$ cm^2 for gold.

From our experimental data we calculated the cross sections for the production of α particles with an energy greater than 20 Mev in bismuth, and the shape of their spectrum. The following results were obtained:

E_p, Mev	130	170*	300	400	480*	530	580	660
$\sigma(E_\alpha > 20$ Mev), 10^{-25} cm^2	0.42	1.03	1.58	1.55	2.03	2.28	1.82	2.1

The method used in the calculation is described in detail in reference 1. The α-particle ranges in the material for energy losses between E + ΔE and E were calculated with the aid of the well-known formulas.[7] The excitation functions of the (α, 2n) and (α, 3n) reactions were taken from the

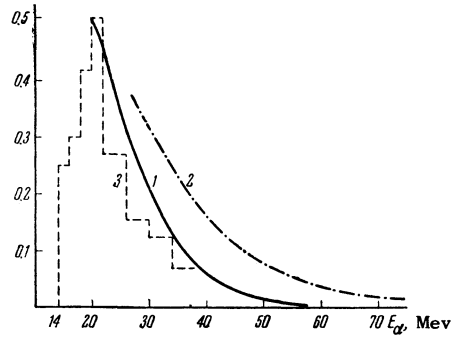
*Corrected results of reference 1.

literature (cf. references 8 — 11). The decreasing portion of the excitation function of the (α, 3n) reaction in the 43 — 60 Mev range was calculated from the formulas of reference 12. The nuclear temperature was taken to be 1.5 Mev, and the neutron binding energy was calculated from the tables in references 13 and 14.*

The shape of the alpha spectrum was taken in the form

$$P(E) = \frac{E-V}{\tau^2} \exp\left(-\frac{E-V}{\tau}\right). \quad (1)$$

The values of the parameters V and τ were chosen to satisfy the experimentally observed mean value of the ratio of the cross sections $\sigma_{At^{210}}/\sigma_{At^{211}} = 0.74$ in the falling form of the spectrum. Such values of the parameter are $\tau = 6$ Mev and V = 12 Mev. The spectrum calculated with these values of τ and V is shown in the figure.



The spectra of the fast α particles (in relative units): 1 — the spectrum calculated from the At^{210} and At^{211} yields from bismuth; 2 — the spectrum calculated from the yields of the thallium isotopes from gold (reference 2); 3 — averaged spectrum of α particles emitted by Bi and W (reference 5).

The same figure shows the spectrum of the α particles produced in the decay of bismuth and tungsten,[5] and the alpha spectrum calculated from the experimental data on secondary reactions in gold.[2] The curves are normalized in such a way that the areas under the curves are proportional to the production cross sections of α particles with corresponding energies.

Our attention is drawn by the difference in the energy spectra of α particles calculated from the secondary-reaction yields in bismuth and gold. In the latter case the spectrum has a smaller slope, and the cross section for the production of α particles turns out to be larger than in bismuth. It seems to us that this discrepancy is due to a possible error in the determination of the relative yields of thallium isotopes because of the difficulty in analyzing the total decay curve of these nuclei. It is impossible to explain this observed discrepancy from the features of the excitation function of the (α, xn) reactions in gold, since a calculation we made with the aid of the formulas of reference 12 showed that the excitation functions for gold are almost the same as for bismuth. However, the authors of reference 2 take account of α particles starting from 27 Mev while the experimental excitation function of the (α, 2n) reaction in bismuth and the calculated excitation function in gold begin at an α-particle energy of ~ 20 Mev.

The production of α particles with an energy larger than 20 Mev cannot be exclusively explained by an evaporation mechanism. Actually, an alpha spectrum satisfying the experimentally obtained ratio of the production cross sections of the two astatine isotopes could only be explained by the evaporation of α particles at an excitation energy of the bismuth nucleus of ~ 600 Mev. Such an excitation energy is unlikely for 600-Mev protons, and completely excluded for protons with lower energies.

On the other hand, the evaporation spectra of α particles with an energy larger than 20 Mev calculated for mean excitation energies of the bismuth nucleus (E_{exc}) due to 120- and 660-Mev protons fall more steeply than was experimentally observed. The spectra were calculated according to formula (1) with $\tau = (cE_{exc})^{1/2}$ ($c = 12.4/A$, A is the atomic weight).[16] The lowering of the Coulomb barrier was taken account of both by means of the formula[16] $V = 0.83V_{class}/(1 + E_{exc}/200)$, and by means of the formula $V = 0.83V_{class}(1 - \tau^2/\tau_c^2)$, where $\tau_c = 9$ Mev.[17] The mean excitation energy of the bismuth nucleus was taken (on the basis of the data of reference 18) to be 190 and 80 Mev for proton energies of 660 and 120 Mev, respectively.

Thus, even at the maximum excitation energies possible in our instance, the obtained spectrum cannot be explained by the evaporation theory.

The fact that the shape of the energy spectrum of the α particles produced in the decay of bismuth nuclei, within the sensitivity of our experiment, does not depend on the energy of the bombarding protons also speaks in favor of this conclusion.

If it is assumed that all 20-Mev α particles are due to evaporation, then it follows from the calculations that on the average half of the α particles with $E_\alpha > 20$ Mev are produced by a different mechanism than the evaporation of α particles

*Cross sections for the capture of α particles by the Bi nucleus were calculated with the aid of a formula proposed by Babykov in reference 15.

from a homogeneously hot excited nucleus. We assume that the fast α particles can be reasonably assumed to have been knocked out from the peripheral region of the nucleus in the process of an intranuclear cascade.[19]

The authors thank B. V. Kurchatov for constant interest in the work and for valuable advice, and V. N. Mekhedov for a number of critical remarks.

[1] Kurchatov, Mekhedov, Chistyakov, Kuznetsova, Borisova, and Solov'ev, JETP **35**, 56 (1958), Soviet Phys. JETP **8**, 40 (1959).

[2] A. E. Metzger and J. M. Miller, Phys. Rev. **113**, 1125 (1959).

[3] Kuznetsova, Mekhedov, and Khalkin, Атомная энергия (Atomic Energy) **4**, 455 (1958).

[4] Batzel, Miller, and Seaborg, Phys. Rev. **84**, 671 (1951).

[5] N. A. Perfilov and V. I. Ostroumov, Dokl. Akad. Nauk SSSR **103**, 227 (1955).

[6] Belyaev, Wang, Sinotova, Német, and Khalkin, preprint, Joint Institute for Nuclear Research, 1959.

[7] B. B. Rossi, High-energy Particles, New York, Prentice-Hall, 1952.

[8] Barton, Ghiorso, and Perlman, Phys. Rev. **82**, 13 (1951).

[9] E. L. Kelly and E. Segrè, Phys. Rev. **75**, 999 (1949).

[10] J. Walter, Phys. Rev. **103**, 704 (1956).

[11] Ramler, Wing, Henderson, and Huizenga, Phys. Rev. **114**, 154 (1959).

[12] J. D. Jackson, Can. J. Phys. **34**, 767 (1956).

[13] A. H. Wapstra, Physica **21**, 367 (1955).

[14] J. R. Huizenga, Physica **21**, 410 (1955).

[15] V. V. Babikov, JETP **38**, 274 (1960), Soviet Phys. JETP **11**, 198 (1960).

[16] K. J. Le Couteur, Proc. Phys. Soc. **A63**, 259 (1950).

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

[18] Metropolis, Bivins, Storm, Miller, Friedlander, and Turkevich, Phys. Rev. **110**, 204 (1958).

[19] V. I. Ostroumov and R. A. Filov, JETP **37**, 643 (1959), Soviet Phys. JETP **10**, 459 (1960).

Translated by Z. Barnea
49