A RADIOCHEMICAL STUDY OF THE (p, pxn), (p, 4pxn), AND (p, 5pxn) REACTIONS

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A radiochemical investigation of the isotopes of bismuth, mercury, and gold produced in the disintegration of bismuth by 660-Mev protons has been carried out. A method for quantitative determination of the radio isotopes, based on L radiation, is proposed for isotopes that have undergone elementary capture. The half-life of Hg^{193} (4 hr) and the E capture in the gold isomer Au^{193m} have been determined by successive extraction of the daughter fraction. The dependence of the yields of the reactions (p, pxn), (p, 4pxn) and (p, 5pxn) on the number of neutrons emitted leads to certain conclusions regarding the mechanism of disintegration of complex nuclei by high-energy protons.

INTRODUCTION

THE reactions (p, pxn), (p, 4pxn), and (p, 5pxn) take place when complex nuclei interact with high-energy protons. The target employed to study these reactions can be an element with only one stable isotope. In the case of medium nuclei, these reactions have been studied for the interaction between protons of various energies with cobalt (60, 100, 170, 240, 370 Mev),[1,2] yttrium (240 Mev),[3] cesium,[4] and tantalum (340 Mev).[5] Whenever several isotopes of the same element could be separated, it was observed that the yield of the (p, 4pxn) and (p, 5pxn) reactions tends to increase as x increases to the maximum, while the yield of the reaction (p, pxn) decreases.

As to heavy nuclei, a study was made of the disintegration of uranium and thorium by 340-Mev protons.[6] The same dependence is observed for these reactions as for light nuclei, but the situation is masked here by the competing fission process, particularly in the case of uranium. The disintegration of bismuth was studied by bombarding it with 375- and 450-Mev,[7] 480-Mev,[8,9] and 660-Mev protons.[9] But in none of these investigations were the yields of more than two or three isotopes of the same element determined. This is explained by the complexity of the decay schemes of heavy nuclei, and also by the lack of a satisfactory procedure for counting the radio isotopes that decay via E capture.

In reference 7, which deals with disintegration of bismuth, the yields of the radio isotopes that disintegrate by E capture have not been corrected for the efficiency of the counting apparatus. In other papers, the count of the radioactivity of the isotopes experiencing capture of an orbital electron, was determined from the K radiation[9] or from the K and L radiation.[8] In either case, to calculate the absolute yields of the above radio isotopes it is necessary to know not only the decay schemes, but also the ratio of the L capture to the K capture, and to take into account the contribution of the soft gamma radiation, the wavelength of which is comparable with that of the K radiation.

It is known[10] that the probability of L capture increases with diminishing transition energy and with increasing forbiddenness of the transition. Theoretical calculations, however, yield a single-valued solution only for the allowed transitions. Since the probability of L_{II} capture in an allowed transition is small, and no L_{III} capture has been observed at all, the ratio of the probabilities of L capture (λ_L) to those of K capture (λ_K) is given by[11]

$$\frac{\lambda_L}{\lambda_K} \sim \frac{\lambda_{L_1}}{\lambda_K} = \left(\frac{W_0 + W_{L_1}}{W_0 + W_K}\right)\frac{g_{L_1}^2}{g_K^2},$$

where g_i is the wave function, W_0 the transition energy, W_i the transition of the i-th level. When $W_0 \gg W_K$ and $W_0 \gg W_{L_1}$, $\lambda_{L_1}/\lambda_K \sim g_{L_1}^2/g_K^2$.

The ratio $g_{L_1}^2/g_K^2$ for allowed transitions, calculated by Rose and Jackson,[12] increases smoothly with increasing mass number and reaches a value of 0.18 for heavy nuclei. At the same time, the experimental L/K capture ratio has a great variety of values for neighboring elements and in some cases, particularly in the range of heavy nuclei, becomes greater than unity.

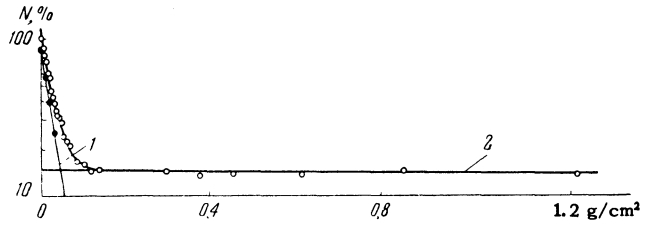

FIG. 1. Absorption curve for electromagnetic radiation from Re^{186} in aluminum: 1) L radiation; 2) gamma background (N is the number of counts as percentage of the initial count).

It follows from the above that at the present time there are not enough theoretical and experimental data on the contribution of L capture in the case of forbidden transitions.

SETUP OF THE EXPERIMENT

In this investigation we studied the reactions (p, pxn), (p, 4pxn) and (p, 5pxn) with radioisotopes of gold, mercury, and bismuth, obtained by disintegrating bismuth with 660-Mev protons. For this purpose, spectrally pure metallic bismuth in quantities of 0.5 or 1 g was irradiated in the internal beam of the synchrocyclotron of the Joint Institute for Nuclear Research. After irradiation, the bismuth was dissolved in concentrated nitric acid and the gold, mercury, and bismuth were separated on suitable isotope carriers. The radioisotopes were identified by their half-lives, by the radiation energy, and by the genetic bonds. The proton current was determined[13] from the yield of the reaction $Al^{27}(p, 3pn)Na^{24}$.

Counting Procedure

To avoid substantial errors due to the indeterminacy in the L/K capture ratio, we determined the activity of the radioisotopes experiencing E capture from their L radiation. This method is effective because K capture is accompanied not only by K radiation but also by L radiation of almost the same intensity. The number of secondary x-ray L quanta that accompany the K capture, can be calculated from the following formula[14]

$$n_L = \omega_K \left(\frac{N_{K_\alpha}}{N_K}\right) + a_K \frac{2(K-LL)+(K-LX)}{\Sigma e_{\text{Auger}}},$$

where ω_K is the fluorescent yield, N_{K_α}/N_K is the ratio of the intensity of the K_α radiation to the intensity of all the K rays, a_K is the number of Auger K electrons, and (K − LX) is the partial yield of the Auger electrons.

The number of secondary L quanta amounts to 80% in the range studied (Z = 80). If the ratio L/K capture ratio is assumed to be unity, the total

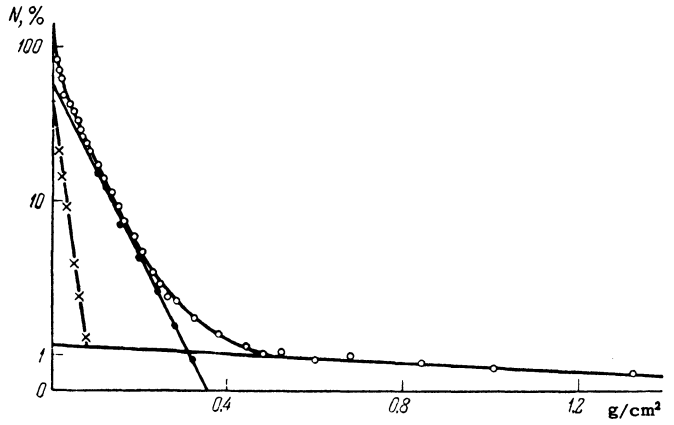

FIG. 2 o) absorption curve of total radiation from Re^{186} in aluminum; ×) L radiation; ●) beta component; solid line – gamma background.

number of L quanta per decay due to E capture amounts to 0.9. Even at very large deviations of L/K from the chosen value, the error in the estimate of the total number of decays deos not exceed 10%.

The detector employed was a standard argon-filled TM-20 end counter which transmitted the K radiation and the soft gamma quanta (0.1 to 0.05 Mev) with almost no absorption (efficiency 0.5%), and which recorded x-rays with an efficiency of 20%. The counter efficiency was calculated from the formula

$$Q = 1 - e^{-\mu d},$$

where d is the thickness of the gas layer of the counter in g/cm^2 and μ is the mass absorption coefficient, found from the tables compiled by Allen.[15] In addition to calculating the efficiency, the counts corresponding to the activity of the radioisotope experiencing E capture were corrected for the following factors: (1) absorption of radiation in the counter window, in the air gap, and in the covering of the sample; (2) self-absorption in the source material; (3) background of hard gamma quanta (determined with the aid of a lead absorber); (4) L-fluorescence yield (according to Burhop's data[16]); (5) number of x-ray L quanta and electrons due to internal conversion (using the decay schemes listed in Table III below. The conversion coefficients were determined from the tables of Sliv and Band[17] and Dranitsina.[18]

The sources used to verify the proposed procedure were the radioisotopes Cu^{64} (K_α radiation of wavelength 1.65 A, close to that of the L radiation from the heavy elements) and Re^{186} (L_α with λ = 1.43 A). The number of E captures or β decays per disintegrating atom of either isotope is found in the literature. We first measured the

TABLE I

Isotope	X-ray wavelength, A	Literature data		Data of present investigation	
		Number of beta decays, %	Number of E captures, %	Number of beta decays, %	Number of E captures, %
$_{29}Cu^{64}$	K_α, $\lambda = 1.65$	58	42 [19]	59	41
$_{75}Re^{186}$	L_α, $\lambda = 1.43$	91	9 [20]	93	7
		95	5 [21]		

total activity with a standard end counter and then deflected the charged particles in the magnetic field and registered only the electromagnetic radiation.

An investigation of the absorption of the radiation from Re in aluminum with the magnetic field on (Fig. 1) and off (Fig. 2) has shown that the absorption curves contain none of the component corresponding to K radiation from tungsten, i.e., the A capture is registered only because of the L radiation. The measurement results listed in Table I are in good agreement with literature data. The accuracy of the method amounts to 20 or 30%, without allowance for the error with which the decay scheme is determined.

Determination of the Half-Life of Hg^{193}

In many cases there were no accurate half-lives available for the mercury radioisotopes. Thus, Seaborg's table[22] lists several values for Hg^{193} (10 hours, 14.5 hours, 29.0 hours, and 5 hours) and none for Hg^{194}. To determine the unknown half-lives we used the Neuman and Perlman method,[23] whereby the daughter fractions are separated from the mother substance at equal time intervals, corresponding approximately to the expected half-life of the mother element. If $N_1\lambda_1$ is the activity of the daughter material and k is the counting efficiency of the daughter material, then

$$kN_1\lambda_1 = N_2\lambda_2 \frac{T_2}{T_2 - T_1}(e^{-\lambda_2\tau} - e^{-\lambda_1\tau}),$$

where $N_2\lambda_2$ is the activity of the daughter material and τ is the growth time. When τ = const, the activity of the daughter substance is proportional to the rate of decay of the mother substance at the start of the period.

To determine more exactly the half-life of Hg^{193}, the daughter gold fractions were separated from the mercury every three hours. The component with half-life of 16 hours belonging to Au^{193}, was separated in the decay curves for gold. The rate of decrease in activity of this component was used to determine the half-life of Hg^{193}, which was found to be four hours (Fig. 3).

To prove that the half-life obtained pertains to the ground state of Hg^{193} and not to the isomer Hg^{193m}, we plotted the theoretical curve for the growth of Au^{193} (T = 16 hr) from Hg^{193} (T = 4 hr), shown in Fig. 1. The data obtained in several experiments are in good agreement with the theoretical values.

E Capture of the Au^{193m} Isomer

It follows from the above that the isomer Hg^{193m} (T = 12 hr), if it is formed at all in the disintegration of bismuth, decays via E capture into the isomer Au^{193m} (T = 3.8 sec), which is directly converted, also by capture of an orbital electron, into the isomer Pt^{193m} (T = 3.8 days), bypassing the ground state of gold (Fig. 5). Indications that the transition $Au^{193m} \rightarrow Pt^{193m}$ is possible are found in the paper by Brunner.[24]

To observe the activity of Hg^{193m}, the daughter platinum was separated from the mercury fraction every four hours. To exclude the influence of the mass-191 chain, the mercury was separated from the bismuth every 12 hours after the termination

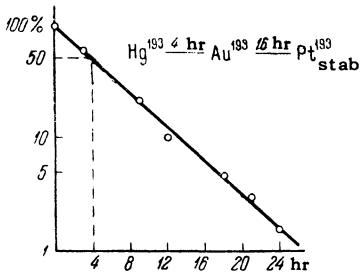

FIG. 3. Radiochemical yield of daughter activity of Au^{193} as a function of time.

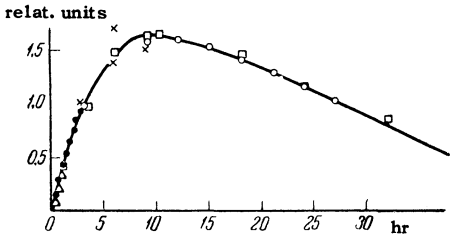

FIG. 4. Growth curve of daughter activity of Au^{193} as a function of time: □) theoretical curve; △, ●, ○, ×) experimental data.

TABLE II

Nucleus	Gamma-transition energy, Mev[26]	Conversion-coefficient ratio, L_{II}/L_{III}[27]	Conversion-coefficient α_{av} (ref. 18)	Transition probability (relative units)
Au^{193m}	0.0324	0.65	17 250	0.03
Au^{195m}	0.0569	1	3 000	0.3
Au^{197m}	0.13	2.5	28	1

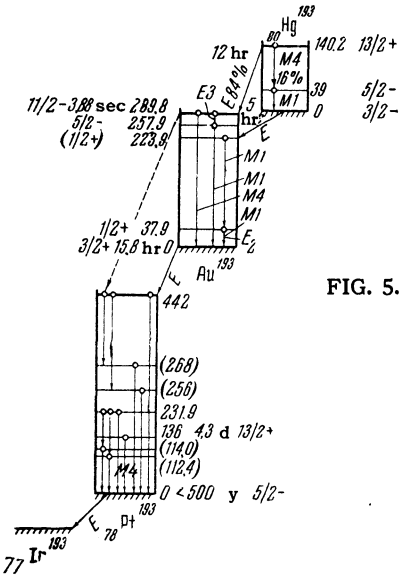

FIG. 5. Decay scheme of Hg^{193}.

of the radiation. From the change of the activity of the platinum (half-life 3.5 days) we found the half-life of Hg^{193m} to be approximately 12 hours. Attempts at observing the three-day activity in the platinum fraction separated from the daughter gold were not successful.

Let us see whether the existence of this transition contradicts the fundamental theoretical premises. The probability of transition of the excited nuclear state for gamma radiation depends on the multipole order of the transition, on the energy of the gamma rays, and on the wave functions of the nuclear states participating in the transition. For specific calculations of the probabilities of the gamma transitions it is necessary to specify some nuclear model. In our case we can consider radiation from an individual proton, moving in the constant field produced by other particles contained in the nucleus. The probability of the electrical 2^L transition can then be expressed by the following Weisskopf formula[25]

$$T_{(EL)} = \frac{4.4(L+1)}{L[(2L+1)!!]^2}\left(\frac{3}{L+3}\right)^2\left(\frac{\hbar\omega}{197\text{Mev}}\right)^{2L+1}$$

$$\times a^{2L} S(j_i, L_{if}) \cdot 10^{21}\ \text{sec}^{-1},$$

Where L is the multipole order, $a = 1.45 \times 10^{-13} A^{1/3}$ cm, S is a statistical factor, and $\hbar\omega$ is the gamma-quantum energy in Mev. The resultant transition probability is corrected for the internal conversion of the gamma radiation:

$$T_{(EL)\,(\text{corr})} = T_{(EL)}(1+\alpha),$$

where α is the internal-conversion coefficient.

It must be noted that the theoretical calculations are rough and cannot be expected to be able to represent exact transitions in actual nuclei. They can serve, however, as the starting point for the interpretation of the experimental results.

We have estimated the probabilities of the gamma transitions from the 11/2 level to the 5/2 level for Au^{193m}, Au^{195m}, and Au^{197m}. The initial calculated results are listed in Table II.

As can be seen from the results, the probability of radiation transition diminishes rapidly from Au^{197m} to Au^{193}. According to the data of Gillion et al.,[28] the probability of decay by isomer transition diminishes also for Hg^{193m}, Hg^{195m}, and Hg^{197m} (in a ratio of 0.16 : 0.5 : 1). At the same time, the probability of E capture increases with increasing neutron deficit of the isotope, i.e., the capture/emission ratio can be rather large for the nucleus considered.

The above data make it possible to conclude that a $Au^{193m} \rightarrow Pt^{193m}$ transition exists and has an intensity of at least 90% (within the limits of experimental accuracy).

Half-Life of Hg^{194}

To observe the activity due to Hg^{194}, the daughter gold was separated from the mercury fraction at various time intervals (from 20 minutes to 20 hours). The 40-hour activity belonging to Au^{194} was found in none of the separated daughter fractions. The mercury was separated from the bismuth within 45 minutes to one hour after the completion of the irradiation. Consequently, the half-life of Hg^{194} is either large or else is less than ten minutes. The latter is not likely, since the Weisskopf mass formula predicts for the $Hg^{194} \rightarrow Au^{194}$ transition an energy less than 80 kev.

A prolonged measurement of the activity of the mercury fraction (on the order of one year) yielded for the long-lived activity a half-life of 130 days.

TABLE III

Isotope	Type of decay	Half-life: From tables of references 18 and 19	Half-life: Our data	$\sigma \cdot 10^{-27}$ cm^2	Decay schemes used, references
$_{79}Au^{191}$	E	3 hr	4 hr	40	[28]
Au^{192}	E, β^+	4,2 "			
Au^{193}	E	15,8 "	16 "	16	[31]
Au^{194}	E, β^+	40 "	39 "	12	[32]
Au^{195}	E	185 d	180 d	1.8	
$_{80}Hg^{190}$	E	90 min	1 hr	12	[33]
Hg^{191}	E	57 "			
Hg^{192}	E, β^+	5,7 hr	5,6 "	22	[28]
Hg^{193m}	E, IT	12 "	11 "	8,3	[33,24]
Hg^{193}	E	5 "	4 "	20	[31,34]
Hg^{194}	E	130 d	130 d	27	[33]
Hg^{195m}	E, IT	40 hr	39 hr	6	[24]
Hg^{195}	E	9,5 "	11 "	12	[32,35]
Hg^{197m}	E, IT	24 "	—	—	—
Hg^{197}	E	65 "	66 "	7	[35]
$_{83}Bi^{201}$	E	2 "	2 "	4	[33]
Bi^{202}	E	95 min		8	[33]
Bi^{203}	E	12 hr	10 "	9	[33,22]
Bi^{204}	E	12 "		21	[33]
Bi^{205}	E	14,5 d	14,5 d	22	[36]
Bi^{206}	E	6,4 "	6,3 "	25	[37]

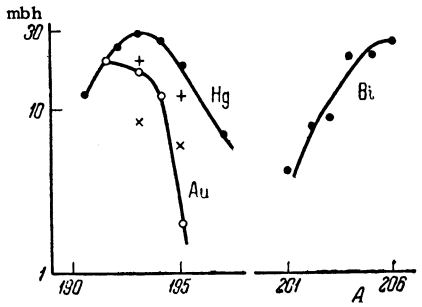

FIG. 6. Dependence of the cross section for the production of radioisotopes of gold, mercury, and bismuth on the mass number, at a proton energy of 660 Mev: +) yield of mercury radioisotopes in the ground state; ×) the same in the isomer state; •) total yield of radioisotopes of mercury and bismuth; o) yield of gold radioisotopes.

This result is in good agreement with the data of Brunner et al.[29] who ascribed the 130-day mercury activity to the mass 194.

During the course of the investigation, we developed chemical procedures that ensure a most exact determination of the separation time of the daughter and mother fractions.* The mercury was separated from the gold by extraction of gold chloride with diethyl ether with subsequent precipitation of the gold with sulfite. The platinum was separated from the mercury and the gold by extraction of chloride complexes of mercury and gold with ethyl acetate, while the platinum remaining in the water film was precipitated with formic acid. Chemical procedures described in reference 30 were used to separate the gold, mercury, and the bismuth from the irradiated bismuth.

DISCUSSION OF THE RESULTS

As a result of the investigation we identified approximately 20 radioisotopes of gold, mercury, and bismuth, produced through (p, pxn), (p, 4pxn), and (p, 5pxn) reactions in the disintegration of bismuth by 660-Mev protons. We also calculated the cross sections for the production of these isotopes. The results are listed in Table III.

The yields of the radioisotopes Hg^{192}, Hg^{193}, Hg^{193m}, Hg^{195}, Bi^{201}, and Bi^{203} have been calculated from the yield of the daughter fractions of platinum, gold and lead. The isomer-pair Hg^{193}/Hg^{193m} and Hg^{195}/Hg^{195m} yield ratio was found to be equal to 2.5. For comparison, we measured the Hg^{193}/Hg^{193m} ratio at a lower bombarding-proton energy. The ratio increases to 8 at a proton energy of 220 Mev, indicating that when the irradiation energy is increased, the yield of the isomer in the higher spin state (Hg^{193m}, spin $^{13}/_2$) is greater than the yield of the isomer in the lower spin state (Hg^{193}, spin $^3/_2$). The results are in good agreement with the data of Hicks and Gilbert.[38]

The dependence of the cross section for the production of the identified isotopes on the mass-number is shown for each element in Fig. 6. As can be seen from the diagram, the course of the curves for mercury and gold, produced via reactions (p, 4pxn) and (p, 5pxn) differs from the curve for bismuth [reaction (p, pxn)]: while the yields of the radioisotopes of gold and mercury increase with the neutron deficit of the isotope and go through a maximum in the case of mercury, the opposite is observed for the bismuth radioisotopes.

To interpret the data obtained it is necessary to consider the mechanism of interaction of high energy particles with complex nuclei. According to the model proposed by Serber,[39] the interaction between a fast proton and a nucleus proceeds in two stages. First the colliding proton knocks out from the nucleus a few fast particles, from essentially neutrons and protons, leaving the remainder of the nucleus in an excited state, which is then deexcited by evaporation of neutrons, protons, and α particles.

The nuclear cascade was first calculated by Goldberger[40] by the Monte Carlo method, and then by a few other authors. The quantitative calculations are laborious and require the use of electronic computers. The calculations show that when 660-Mev protons interact with a bismuth nucleus,

*L. V. Filatova helped with the chemical operations.

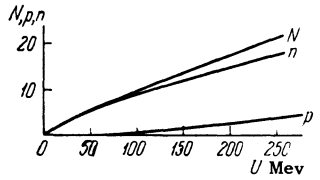

FIG. 7. Number of evaporated particles as a function of the excitation energy U: N) total number of nucleons, n) neutrons, p) protons.

of two fast protons and two fast neutrons are emitted on the average from the latter.[41]

The evaporation of the excited nucleus is satisfactorily explained with the aid of nuclear thermodynamics. In our case, without introducing a substantial error, we can consider the evaporation of neutrons and protons only. Then, according to the simplest variants of nuclear statistical theory, the total number of evaporated nucleons can be calculated with the aid of the following equations:[9]

$$-\frac{\partial U}{\partial (n+p)} = \frac{T_n (E_n + 2T) + T_p (V_p + E_p + 2T)}{\Gamma_n + \Gamma_p},$$

$$\frac{\Gamma_p}{\Gamma_n} = \left(1 + \frac{V_p}{T}\right) e^{-\frac{V_p}{T}} \frac{Z}{A-Z},$$

where U is the excitation energy of the nucleus, γ_n and γ_p are the widths of the energy levels of the neutrons and protons (proportional to the emission probability), E_x is the particle binding energy in the nucleus ($E_p = E_n = 7.5$ Mev), A is the mass number, C is the nuclear charge, E_p is the height of the Coulomb barrier for protons (8 Mev), and T is the temperature of the nucleus ($0.22\sqrt{U}$).

The calculated dependence of the total number of evaporated particles N, the number of the neutrons n, and the number of protons p on the excitation energy is shown in Fig. 7. If the knock-out component contains two protons and two neutrons, the evaporation involves, two protons and seven to eleven neutrons in the case of gold, and two protons and six to thirteen neutrons in the case of mercury. The greatest yield, according to our measurements, is obtained with the radioisotope Hg^{193} (two protons and eleven neutrons are evaporated), corresponding to a most probable value of excitation energy of 150 Mev. This value may be somewhat too high, if the yield of the radioisotopes of thallium, which were not investigated in this work, is greater than that of the mercury radioisotopes. The most probable excitation energy is then reduced to 100 or 110 Mev.

When bismuth nuclei are formed, there is emitted from the nucleus one proton that can be attributed with full justification to the knock-out component. In this case neutrons alone will be evaporated from the nucleus and, according to Fig. 7, the most probable excitation energy should not exceed 50 Mev.

Our experimental results are in good agreement

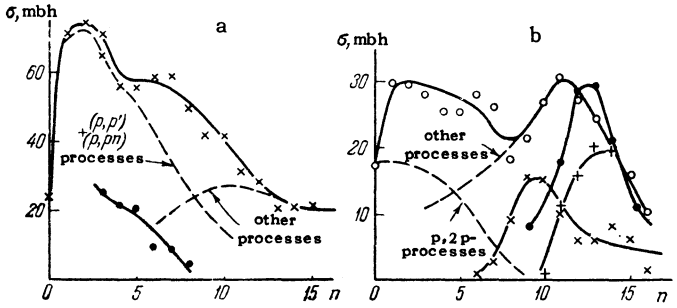

FIG. 8. Cross sections for disintegration as a function of the number of emitted neutrons n and the number of protons p: a) for p = 1 (× – calculated values, ● – experimental values for Bi); b) for p = 2 (○ – calculated values, ● – experimental data for Hg) and for p = 3 (× – calculated values, + – experimental data for gold).

with the theoretical calculations of Jackson.[42] The cross sections he calculated for the disintegration of elements with A ~ 200 by 400-Mev protons are shown in Figs. 8A and 8B, which also show, for comparison, our experimental data.

It is seen from Fig. 8 that the curve for the bismuth isotopes is similar to the theoretical curve, and that the knock-out mechanism is decisive for the (p, pxn) reaction. The discrepancy in the absolute values of the yields of the (p, pxn) reaction can apparently be ascribed to the fact that our experimental data have been obtained at a higher proton energy.

The dependence of the yields of the reactions (p, 4pxn) and (p, 5pxn) (Fig. 8B) is in good agreement with the theoretical data for 2p and 3p. This fact can be readily explained by the circumstance that when he calculated the evaporating process, Jackson[42] neglected the evaporation of protons, while it is seen from Fig. 7 that on the average 10 to 12 evaporated neutrons are associated with two evaporated protons. It follows from the comparison of the theoretical and experimental values that the evaporation process plays the decisive role in the formation of radioisotopes of mercury and gold and dictates the character of the dependence of the radioisotope yields on the mass number.

The same characteristic features of the reactions (p, pxn), (p, 4pxn), and (p, 5pxn) are also observed in the disintegration of elements of medium atomic weights,[1-5] since the difference in the yields of neighboring isotopes of the same element increase with diminishing Z of the irradiated nucleus and with diminishing energy of the incident protons.

It can thus be concluded that in the disintegration of complicated nuclei by high-energy protons,

the distribution of the residual nuclei is caused by the following interaction mechanisms: (1) during the course of the reactions (p, 4pxn) and (p, 5pxn), the decisive process is evaporation and the yield of the radioisotopes increases with increasing neutron deficit and goes through a maximum; (2) the (p, pxn) reactions occur when a small excitation energy is transferred to the nucleus, the emission of a large number of neutrons is difficult, and consequently the yield of the radioisotopes diminishes with increasing neutron deficit.

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