SECONDARY CAPTURE OF LITHIUM NUCLEI BY LEAD

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The formation of $At^{211,210,207}$ in lead under bombardment by 80 — 660 Mev protons, 75 — 370 Mev deuterons and 210 — 810 Mev alpha particles has been studied by radiochemical means. The astatine isotopes result from secondary capture of lithium nuclei produced through disintegrations and having kinetic energies exceeding the Coulomb barrier. The At^{211} yield under alpha-particle bombardment reaches 0.3 microbarn and is practically independent of the alpha-particle energy. Under proton and deuteron bombardment the yield increases with particle energy, especially when the proton energy exceeds 400 Mev, and attains 0.2 microbarn at 660 Mev. The At^{211} yield is independent of lead target thickness in the 0.3 — 1.6 mm range and decreases for thicknesses smaller than 0.3 mm. The production cross section for the captured lithium fragments is computed and their energy spectrum is estimated on the basis of the astatine yield from lead. The cross section for production of "over-Coulomb" lithium fragments by 660-Mev protons is 3 — 6 millibarns.

INTRODUCTION

THE most interesting aspect of the fragmentation process,[1-6] by which we mean the ejection of lithium, beryllium and heavier fragments from excited nuclei, is the emission of fragments having energies that exceed the Coulomb barrier. No satisfactory theoretical explanation has thus far been advanced for this phenomenon. None of the known mechanisms of nuclear reactions can account for the fact that an aggregate of nucleons may, without being disrupted, receive an amount of kinetic energy which sometimes exceeds the total binding energy of the nucleons in the fragment.

Radiochemical investigations of "secondary" reactions[7-11] provide one method for the study of this process. These secondary reactions are produced in nuclei of the target by secondary "over-Coulomb" fragments. In the present work we have studied the formation of astatine isotopes in the secondary reaction $_{82}Pb(Li, xn)_{85}At$ when lead is bombarded with high-energy protons, deuterons or alpha particles. Lead was chosen as the target material for two reasons. The undesirable bismuth, uranium and thorium impurities can be removed relatively easily from lead. Also, despite the very low reaction yield (cross section) of $10^{-30} — 10^{-32}$ cm^2 the astatine end-product can be detected conveniently by means of its alpha emission. Chemical removal of beta- and gamma-active contamination from the reaction products is considerably simplified.

EXPERIMENTAL PROCEDURE

For reliable observation of a secondary reaction, the lead target must not contain more than 10^{-3}, 10^{-4} or 10^{-5}% of bismuth, uranium, or thorium, respectively.[10] This level of purity was attained as follows. The original chemically pure lead carbonate was used to prepare lead nitrate, which was recrystallized twice from a 75% solution (by volume) of methyl alcohol and once from concentrated nitric acid. The nitrate was heated and the resulting lead oxide was reduced to the metal by means of sucrose at 700 — 800° C. The original lead carbonate contained 10^{-2}% bismuth, but the lead metal revealed no trace of bismuth ($< 10^{-3}$%).* Uranium and thorium impurities in the metallic lead were estimated from the Ra^{223} yield under bombardment by 120-Mev protons and amounted to $< 10^{-5}$% if we assume a ~10-millibarn cross section for the formation of Ra^{223} from these elements.[12]

The targets were bombarded with 80 — 660 Mev protons, 75 — 370 Mev deuterons and 210 — 810 Mev alpha particles. The bombarding energy was varied by placing the target at different radial dis-

*Bismuth impurity in the lead was determined spectroscopically by M. Farafonov of the GEOKhI (Institute of Geochemistry and Analytical Chemistry), to whom the authors wish to express their appreciation.

tances in the beam path. In order to obviate the loss of astatine through target heating by the proton or deuteron beam, the lead samples, which weighed about 1 gram, were sealed in quartz ampoules with an outside diameter of 4 mm, 30 mm length and 0.5 — 0.6 mm wall thickness. Irradiation periods varied from 0.2 to 2 hours.

For the purpose of determining the astatine yield, 660-Mev protons were used to bombard lead foils of different thicknesses placed on the end faces of plates forming the magnetic extracting channel[13] of the synchrocyclotron. The proton beam was greatly diffused at the plates and the flux was attenuated by a factor of 50 — 100 compared with the circulating beam. All of the foils (each measuring 3 × 40 mm) were placed in a row in a single plane perpendicular to the proton beam and were bombarded simultaneously during 2 — 10 hours.

Diisopropyl ether was used to extract astatine from the irradiated lead dissolved in hydrochloric acid. For further purification, the radioactive impurities were coprecipitated with elemental tellurium from an alkaline solution, and the astatine was coprecipitated with elemental tellurium from a hydrochloric acid solution. (Details of the chemical technique for separating astatine are given in reference 14.) As a control some lead samples were treated by the procedure described in reference 10, which is based entirely on the coprecipitation of astatine with tellurium; the astatine yields agreed, within experimental error, with those of the extraction procedure.

Our measuring technique and apparatus have been described in reference 15. In all experiments both 7.5 -hour and ~ 140-day alpha activity were detected, which we assigned to At^{211} and Po^{210}. In some experiments activity with a half-life of about 2 hours was observed and was assigned to At^{207}.

The intensity of the bombarding beam was determined through the yield of Na^{24} from the aluminum foil in which the samples were wrapped during irradiation. The technique for measurements on Na^{24} was the same as that described in reference 15. The cross sections for N^{24} formation from Al^{27} under different bombarding energies was taken from references 16 — 19. For deuterons with > 200 Mev and alpha particles with > 400 Mev, the cross sections for Na^{24} formation were determined by extrapolating the excitation curves of Na^{24} yield given in references 16 and 17. In the case of deuterons this cross section was taken to be 22 millibarns, while for 585- and 810-Mev alpha particles it was estimated at 18 and 13 millibarns, respectively.

Possible errors in beam monitoring during irradiation of the ampoules were determined by comparing the cross sections for astatine formation at 660 Mev, in the case of irradiation at the magnetic channel with and without ampoules and in the circulating beam at reduced intensity. The yields in all three cases agreed within experimental error.*

EXPERIMENTAL RESULTS

The yields of At^{211} and At^{210}, and the relative yield At^{207}/At^{211} at different proton energies, are given in Table I, where (as everywhere below) the averages of at least three determinations are given. A single determination was obtained only in the case of ~ 80-Mev protons. Random errors for the astatine yield given in Table I do not exceed ±30%. The relative yield of At^{207} takes the 90% K-capture branching fraction into account.

TABLE I

Proton energy, Mev	Yield (cross section), microbarns		At^{207}/At^{211}	Total yield (cross section), microbarns
	At^{211}	At^{210}		
660	0.17	0.21	~1.3	~1.3
500	0.06	0.10	–	~0.35
340	0.03	0.08	–	~0.2
120	0.005	0.01	~1.1	~0.03
~80	~0.01	–	–	–

$At^{205}(\alpha, K)$ with $T_{1/2}$ = 25 min was detected in one run with 660-Mev protons, and its relative yield was estimated. When K capture is neglected we have $At^{205}/At^{211} \sim 0.1$.

The At^{211} and At^{207} yields are almost identical for 660- and 120-Mev protons. For 500 — 120 Mev, the At^{210} yield is about twice as large as that of At^{211}. The yields of the different astatine isotopes decrease with decreasing proton energy in approximately the same manner. The At^{210}/At^{211} ratio averaged over all proton energies is 1.5 ± 0.5. The last column of Table I gives the total yield of isotopes from At^{207} to At^{211}. The yields of At^{209} and At^{208} were interpolated from the yields of At^{211}, At^{210}, and At^{207}. Random errors included in the total yield do not appear to exceed ±50%.

The At^{211} yield at different proton, deuteron, and alpha-particle energies are shown in Fig. 1.

*In the present work all targets were thick enough to make the yield independent of target thickness, with the exception of the experiments which were specifically intended to determine the dependence on target thickness.

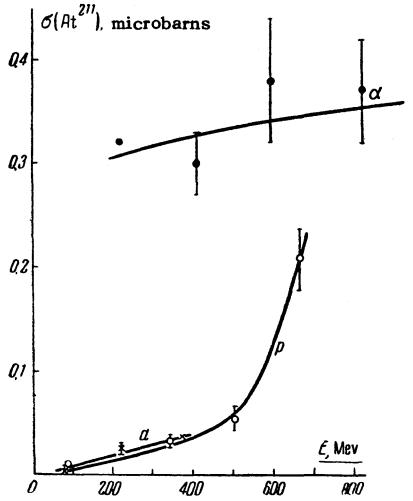

FIG. 1. At^{211} yield as a function of bombarding energies for alpha particles (α), deuterons (d) and protons (p).

The highest At^{211} yield (~ 0.3 microbarn) was detected under alpha-particle bombardment and varies only slightly with increasing alpha-particle energy. For deuterons and protons up to 400 Mev the At^{211} yields are almost identical, and are approximately one-tenth as large as for alpha-particles. Above 400 Mev, protons produce a strong increase of the At^{211} yield, which for 660-Mev protons becomes more than half of that for alpha particles.

It should be noted that in the case of high-energy alpha particles At^{211} can also be formed through alpha-particle capture by Pb^{208} followed by π^- and neutron emission, and also through capture by Pb^{207} followed by π^- emission. However, the figure shows that these reactions, if they do occur, are weak and not decisive.

Figure 2 shows the At^{211} yield from lead foils of different thicknesses. Almost no change is observed in the range 0.3 — 1.6 mm. Below 0.3 mm the yield decreases gradually and for 0.03 mm it amounts to half of the plateau value.

DISCUSSION

The observed quantity of astatine could not have been formed through the disintegration of any possible uranium, thorium, or bismuth impurities. It is estimated in the case of 120-Mev protons that these impurities could not account for more than one-tenth of the At^{211} and At^{210} yields. For protons with energies above 120 Mev this fraction becomes even smaller. The observed astatine activity therefore results almost entirely from the secondary reaction of lithium-capture reaction.

In Fig. 3 the proton-energy dependence of the total yield of astatine from lead is compared with an analogous curve for the formation of iodine from tin.[9] The curves are similar and at 660 Mev the astatine and iodine yields coincide.

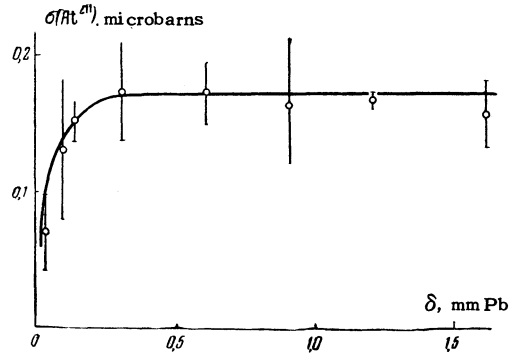

FIG. 2. At^{211} yield from lead foils of different thicknesses δ compared with a calculated curve.

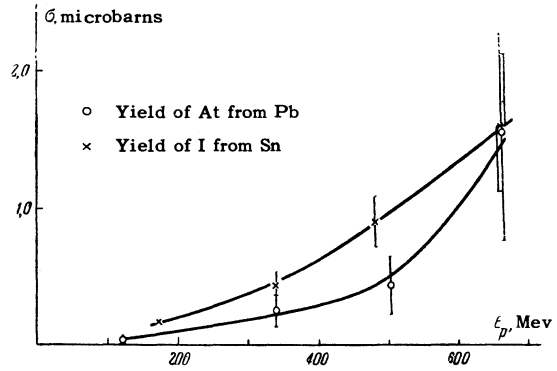

FIG. 3. Total cross section for the formation of astatine isotopes from lead and of iodine isotopes from tin as a function of proton energy.

The total astatine yield (~ 0.2 microbarn) which we obtained with 340-Mev protons agrees satisfactorily with the analogous yield of lead from gold (0.36 microbarn) obtained with 380-Mev protons.[11]

The procedure described in reference 10 was used in conjunction with the yields of secondary-reaction products to compute the energy spectrum of the lithium fragments and the cross sections for their formation from lead under bombardment by high-energy protons. The experimental yield ratios At^{210}/At^{211} and At^{207}/At^{211} had to follow from the selected lithium spectrum in conjunction with the given excitation functions for the reactions Pb (Li, xn) At (x = 1, 2, 3, ..., 8) and the known losses of energy through ionization. The spectrum was then used to compute the cross section for lithium formation. On the basis of the data in reference 5 the lithium fragment spectrum was represented by*

$$P(E)\,dE = \tau^{-2}(E - V)\,e^{-(E-V)/\tau}\,dE \qquad (1)$$

with suitable values of the parameters V and τ.

*We attach no physical significance to this shape of the energy spectrum. $P(E)dE = E^{-h}dE$, with $h \approx 2$, could also be used, but the selected formula furnishes a somewhat better approximation of all known experimental data.

It was assumed that the energy spectra for Li^6, Li^7, and Li^8 are identical.[5] The excitation functions for the capture of lithium fragments by lead isotopes were calculated separately for Li^6, Li^7, and Li^8 by means of Jackson's formulas,[20] while Babikov's formula was used to calculate the cross section for lithium capture by lead.

Figure 4 shows the calculated excitation functions for the principal reactions in the formation of At^{211}, At^{210}, and At^{207} when Li^6 and Li^7 are captured by different lead isotopes. 25 out of 43 possible reactions were considered. In each instance we took into account the abundance of the lead isotope, and the relative yields of Li^6, Li^7, and Li^8 from lead were taken to be 0.55 : 0.41 : 0.043 as in the case of gold.[11] Familiar formulas[22] were used to calculate the ranges of different fragments in lead.

Table II gives the calculated relative yields

TABLE II

V		At^{210}/At^{211}	At^{207}/At^{211}	σ_{Li}, mb
6	11,5	1,67	0,9	6,0
	10,5	1,63	0,79	6,1
	9,5	1,57	0,63	6,7
10	11,5	1,7	0,86	5,7
	10,5	1,65	0,8	6,0
	9,5	1,57	0,63	6,7
15	6,5	1,39	0,28	9,4
	5,5	1,27	0,15	11,4
	4,5	1,16	0,09	14,5

At^{210}/At^{211} and At^{207}/At^{211} together with the cross section for the formation of lithium fragments with > 30 Mev in the case of 660-Mev protons, for different values of V and τ. The critical quantity determining the energy spectrum is seen to be the relative yield At^{207}/At^{211}, whereas At^{210}/At^{211} and $\sigma_{Li}(E_{Li} > 30$ Mev) change very little as V and τ are varied. V = 15 Mev and τ = 4.5 — 6.5 Mev correspond to relative yields At^{207}/At^{211} that differ strongly from the experimental results (Table I). V = 6 — 10 Mev and τ = 10.5 — 11.5 Mev in (1) furnish the energy spectra that best satisfy all experimental data, including those in the literature. Figure 5 shows the spectrum of lithium fragments with energies above 30 Mev (τ = 11.5 Mev, V = 6 Mev). Satisfactory agreement is found between the shape of our spectrum and the calculated spectrum of lithium fragments from secondary reactions in gold.[11] However, the cross section for the formation of lithium nuclei with > 54 Mev from gold was four times as large as our result obtained with 340-Mev protons (σ_{Li} = 1 mb from gold; 0.27 mb from lead).

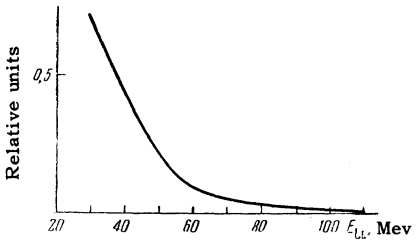

FIG. 5. Spectrum of lithium fragments with energies >30 Mev, calculated from the secondary-reaction yield in lead.

The approximate constancy of the relative yield At^{207}/At^{211} for 660-Mev and 120-Mev protons (Table I) indicates that the spectrum of over-Coulomb lithium fragments is either independent of proton energy or is only very slightly dependent. As a check, the calculated probability of At^{211} formation as a function of lead-foil thickness was compared with the experimental curve. For the purpose of the calculation we estimated the effective ranges of lithium nuclei in foils of different thicknesses, using the angular distributions of fast lithium fragments given in reference 5. The probability of At^{211} formation in lead of thickness δ was computed as the difference between the probability w_0 of formation by a fragment of the given energy in infinitely thick lead and the probability w of formation by a fragment with this same amount of energy remaining after it had traversed a lead layer of thickness δ. We have

$$w_0 = \sum_{i=1}^{n} N_0 \sigma_i \Delta l_i, \qquad (2)$$

where N_0 is the number of lead atoms per cm^3,

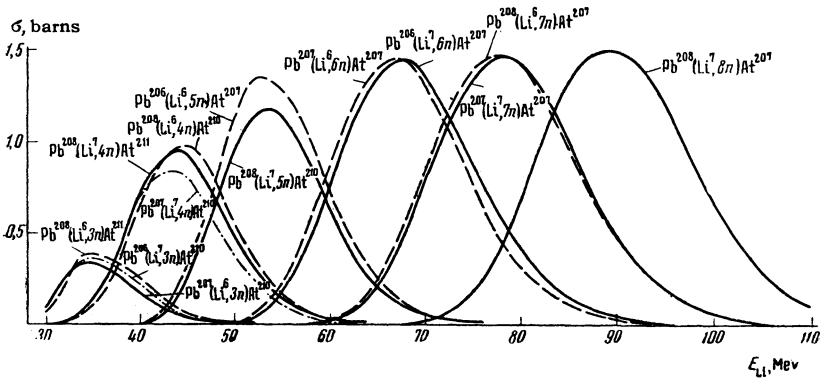

FIG. 4. Excitation functions for the principal reactions in the formation of $At^{211,210,207}$ when Li^6 and Li^7 are captured by different lead isotopes.

σ_i is the cross section for lithium capture leading to At^{211} formation, Δl_i is the ionization range of a fragment in lead within the energy range $(E_i + \Delta E_i; E_i)$. The probability of At^{211} formation was integrated numerically over the entire lithium energy spectrum for each thickness δ.

The curve in Fig. 2 represents the calculated dependence of the probability of At^{211} formation; agreement with experiment is found at thicknesses beginning with 0.24 mm. The curve satisfactorily represents the falling-off of experimental values for small lead thicknesses. The postulated identity of the spectra of different lithium isotopes and the assumption of a value for the $Li^6 : Li^7 : Li^8$ ratio, as well as the possible differences between the calculated and actual excitation functions, somewhat reduce the reliability of the conclusions reached with respect to both the spectrum and the dependence of the yield on foil thickness. These conclusions must therefore be regarded as only qualitative.

Our experimentally observed dependence of the probability of At^{211} formation on lead-foil thickness permits an independent estimate of the cross section for the production of over-Coulomb lithium nuclei. This cross section, which we denote by σ^p_{Li}, is obtained from the relationship

$$B = N_0 \sigma^p_{Li} \overline{\sigma^{Li}_{At}} \Delta l. \quad (3)$$

Here B is the astatine yield for a given proton energy, $\overline{\sigma^{Li}_{At}}$ is the energy-averaged cross section for lithium capture by lead isotopes with subsequent astatine (At^{211}) formation. B and Δl are obtained directly from Fig. 2: B is the ordinate of the curve at saturation, and Δl represents roughly half of the lead thickness at which the probability of At^{211} formation begins to decrease from saturation. $\overline{\sigma^{Li}_{At}} = 0.1$ barn was calculated from the excitation functions for the reactions resulting in At^{211} formation, taking into account the abundances of the lead isotopes and the yields of different lithium nuclei. Substitution into (3) gives σ^p_{Li} 3 — 4 millibarns, which agrees satisfactorily with the cross section for the formation of over-Coulomb lithium fragments that is calculated from the energy spectra.

Some remarks on the mechanism for the formation of over-Coulomb fragments are in order. Although certain authors have used the statistical model[23,24] to account for various characteristics of fragmentation, this model cannot account for the origin of over-Coulomb fragments. The statistical model accounts for most of the fragments having energies close to that of the Coulomb repulsion, but not for the considerable fraction of over-Coulomb fragments. This can be seen from reference 4, for example, where emulsions were used to study the energy spectra of fragments. The authors of this paper discuss the partial success of evaporation theory and point to the need for some new mechanism. Our own work also shows that the energy spectrum of over-Coulomb fragments cannot be explained by means of the statistical model. If the parameter $\tau = 10.5 — 11.5$ Mev is given the physical meaning of a nuclear temperature, as is required in evaporation theory, we arrive at the absurd result that the excitation energy of the nucleus is several times greater than the energy of the bombarding particles. On the other hand, when we insert into (1) the values V = 15 Mev and $\tau = 4.5 — 5.5$ Mev, which are reasonable from the point of view of evaporation theory, we see from Table II that At^{207}/At^{211} is considerably below the experimental value.

The formation of over-Coulomb fragments cannot be accounted for within the statistical model by any possible "local" superheating of a nucleus as a result of pion creation and absorption, for example.[25,26] Figure 1 shows that over-Coulomb fragments are also produced at bombarding energies lying considerably below the threshold for meson production.

Over-Coulomb fragments are in all likelihood ejected before nuclear heating occurs, so that the statistical treatment is altogether unsuitable. This is indicated, for example, by the relationship between the yield of high-energy fragments and the number of cascade particles in disintegrations,[4] as well as by the high degree of forward emission of over-Coulomb fragments which has been noted in almost all studies of fragments in emulsions. We consider it more promising to regard the formation of over-Coulomb fragments as the result of direct many-particle interactions between nucleons of the target nucleus and both the bombarding particle and cascade nucleons.[27] Such interactions seem possible if we assume that the distance of nucleon separation in nuclear matter fluctuates so that nucleons can briefly come much closer than the average distance. Under these conditions an incoming particle could interact with a fluctuating group of nucleons as a whole and transfer to the latter a considerable fraction of its energy. However, we do not believe that we can apply the fluctuating-compression model of nuclear matter in the form that has been developed to account for the emission of high-energy deuterons.[28] This model yields a very small probability for the formation of over-Coulomb fragments and does not at all account for the fact

that we have observed the fragment yields to depend differently on proton and alpha-particle energies.

It must be noted that if many-particle interactions are responsible for the formation of over-Coulomb fragments, then reactions with multiply-charged ions should produce instances of the inverse event wherein the energy of an incoming ion is transferred to single nucleons. The fact that Karamyan and Pleve[29] have observed events in which the entire excitation energy (~ 60 Mev) was transferred to two nucleons indicates that such inverse many-particle interactions may occur.

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