

STUDIES OF THE SECONDARY REACTION OF LI-CAPTURE IN LEAD 1960, + 39, 63, c 527-535. nezia

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STUDIES OF THE SECONDARY REACTION OF LI-CAPTURE IN LEAD

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### Summary

Production of astatine isotopes (At<sup>211</sup>, At<sup>210</sup>, At<sup>207</sup>) in lead bombarded with 80-660 MeV protons, with 75 -370 MeV deuterons and with 210-810 MeV

 $\bigwedge$ -particles have been studied by the radiochemical method. These isotopes of astatine are a result of the secondary reaction of over Coulomb Li-oapture which is obtained in fragmentations. The yield of At<sup>211</sup> bombarded with  $\bigwedge$ -particles amounts to 0.3 microbarn and practically does not depend on the energy of

 $\checkmark$ -particles. When bombarded with protons and deuterons the yield rises with increasing energy and especially when bombarded with protons of energy higher than 400 MeV and reaches 0.2 microbarn at 660 MeV. The yield of At<sup>211</sup> with respect to the thickness of a lead target is constant within 0.3 - 1.6 mm and decreases for targets less than 0.3 mm. The formation cross section and the energy spectrum of captured Li fragments have been estimated by the yields of astatine isotopes from lead. The formation cross section of over Coulomb Li fragments is 3-6 millibarn at 660 MeV.

## Introduction

In the process of fragmentation  $^{1-6/}$ , which is understood as the ejection of Li, Be and heavier fragments by an excited nucleus, the ejection of over Coulomb barrier fragments is of special interest. This phenomenon has no satisfactory theoretical explanation up to now. The transfer to a nucleon complex of so a large kinetic energy which exceeds sometimes the total coupling energy of nucleons in a fragment cannot be accounted for by any known mechanism of nuclear reactions (without destroying that complex).

Radiochemical study of 'secondary reactions'<sup>/7-11/</sup> is one of the methods for investigation of this process. Reactions taking place in the nuclei of the target material induced by the secondary over Coulomb barrier fragments are usually called secondary reactions. This paper describes production of astatine isotopes according to the secondary reaction  $Pb_{82}(Li, xn)_{85}$  At (where x is the number of ejected neutrons) when lead is bombarded with high energy protons, deuterons and  $\checkmark$  -particles. The choice of lead as an object for bombardment is caused by two reasons. On one hand Bi, U and Th are dangerous contaminations in case with lead. The purification from these elements is achieved comparatively easily. On the other hand in view of the expected negligibly small effects ( the yield of the reaction  $10^{-30} - 10^{-32} \text{ cm}^2$ ) the final product of the reaction-astatine is more conveniently observed by the emitted  $\backsim$  -rays. In this case the requirements for chemical purification of the studied products of the reaction from the disturbing  $\beta$ ,  $\beta$  -radioactive contaminations are considerably simplified.

## Experimental Procedure

For observation of the secondary reaction Pb should not contain Bi, U and Th contaminations above 10<sup>-3</sup>%, 10<sup>-4</sup>% and 10<sup>-5</sup>%, respectively/10/. The indicated degree of lead purity was achieved as follows: chemically pure carbonate served as an initial product. Nitrate prepared from it was twice crystallized from 75% solution ( by volume) of methyl alcohol and once from the concentrated HNO<sub>3</sub> acid. Nitrate was ignited to oxide. Lead oxide was reduced to metal with sacharose at 700-800° C. The content of Bi in initial lead carbonate was  $10^{2}$ %. Bi has not been found in metallic lead (  $< 10^{-3}$ %)\*. U and Th contamination in metallic lead was estimated by the yield of Ra<sup>223</sup> when lead was irradiated with 120 MeV protons. U and Th contamination in lead has been found to be  $< 10^{-5}$ %. The cross section of Ra<sup>223</sup> production from the above elements is  $\sim 10$  millibarn/12/.

The targets were bombarded with 80-660 MeV protons, with 75-370 MeV deuterons and with 210-810  $\checkmark$  -particles. The change of the incident particle energy was achieved by locating the target at different radii of the particle beam orbit. In order to avoid the losses of astatine in heating the targets with proton and deuteron beams, the samples of lead about 1 gr were soldered in quartz ampules. The quartz ampules had the following dimensions: length - 30 mm, the outer diameter - 4 mm, wall thickness-0.5-0.6 mm. The periods of irradiation lasted from 0.2 up to 2 hours.

In experiments on determination of the yield of cstatine from lead foils of different thickness the samples were irradiated at surfaces of the plates of the synchrocyclotron magnetic channel/13/ with proton energy of 660 MeV. The proton beam at the end of the magnetic channel plates was considerably blown up and had the intensity per  $1 \text{ cm}^2$  50-100 times smaller than that of the circulating beam. In this case the whole set of foils ( $3 \times 40 \text{ mm}$  each ) was exposed simultaneously. The foils were placed near each other in the same plane which was perpendicular to the proton beam. The duration of irradiation in these experiments was 2-10 hours.

Astatine was released from the irradiated lead by extracting it from the hydrolic acid solution by use of disopropyl ether. Further refinement of the preparation was performed by coprecipitating of radioactive contamination with elementary tellurium and from hydrolic acid solution. The detailed method of chemical isolation of astatine is given in/14/. As a test of checking several samples were treated as described in /10/. The method described was based entirely on coprecipitating of astatine with tellurium. The yields of astatine in these cases were obtained the same within experimental errors as by the extraction method.

The method for measurement of samples and the employed technique are described in<sup>15/</sup>. In all the experiments  $\checkmark$ -activity was found to have the half-life of 7.5 hour and  $\sim$ 140 days which we assigned to At<sup>211</sup> and Po<sup>210</sup>. In some of the experiments we found the activity with the half-life of  $\sim$ 2 hour ascribed to At<sup>207</sup>.

\* The authors are grateful to M.Farafonov, a member of the G.E.O.K.I. research staff, who has performed spectral determination of bismuth contamination in lead.

The beam intensity of the bombarding particles was determined by the yield of Na<sup>24</sup> from the alluminium foil in which the samples were wrapped while exposing them. The method for measuring the preparation of Na<sup>24</sup> was the same as in/15/. The formation cross section of Na<sup>24</sup> from Al<sup>27</sup> at different energles of bombarding particles were taken from/16-19/. For deuterons of energies higher than 200 MeV and for  $\checkmark$  -particles of energies higher than 400 MeV the formation cross sections of Na<sup>24</sup> were determined by extrapolating the curves/16-17/. In the case with deuterons this cross section was taken to be 22millibarn. For  $\checkmark$  -particles of 585 and 810 MeV it was evaluated to be equal to 18 and 13 millibarn, respectively.

Possible errors in monitoring the beam in irradiating the samples in the ampules were determined by comparing the cross section of astating formation at 660 MeV when bombarded in an ampule and without it at the magnetic channel and also at the circulating beam with reduced intensity. In all three cases the obtained yields of the reactions\* are in agreement within experimental errors.

# Experimental Results

Table 1.

Proton energy MeV	Yield (m <sub>At</sub> 211	icrobarn) At <sup>210</sup>	At <sup>207</sup> At <sup>211</sup>	Total yield (microbarn)	
660 500 340 120	0.17 0.06 0.03 0.005	0.21 0.10 0.08 0.01	~1.3 ~ 1.1	1.3 0.35 0.2 0.03	
80	0.01	•••		a and a second s	

\* In this investigation all the irradiations were carried out at 'thick' targets in which the yield of the reaction does not depend on the thickness of the sample. Experiments on the determination of the yield dependence on the target thickness are an exception.

It is seen from the table that the yields of  $At^{211}$  and  $At^{207}$  are nearly equal at incident protón energies of 660 MeV and 120 MeV. The yield of  $At^{210}$  at 500-120 MeV is approximately two times greater than the yield of  $At^{211}$ . With decreasing proton energy the yield of astatine isotopes decreases approximately equally. The relative yield averaged at all energies of incident protons is equal to  $1.5 \pm 0.5$ . The last column of Table 1 presents the total yield of isotopes from  $At^{207}$  up to  $At^{211}$ . The yields of  $At^{209}$  and  $At^{208}$  are interpolated by the yields of  $At^{211}$ ,  $At^{210}$  and  $At^{207}$ . An accidental error in the determination of the total yield of astatine does not exceed, apparently,  $\pm$  50%.

The yield of the isotope  $At^{211}$  at different proton, deuteron and  $\alpha$ -particle energies is shown in Fig. 1. As is seen in the figure the largest yield of  $At^{211}$  is observed in bombardment with

Besides it should be noted that when the energy of  $\propto$ -particles is high, the isotope At<sup>211</sup> can be formed in the reaction of  $\propto$ -particle capture in the isotope Pb<sup>208</sup> with the following emission of a  $\pi$ -meson and a neutron and also in the reaction of the particle capture in Pb<sup>207</sup> with the emission of a  $\pi$ -meson. However, as is shown in the figure the contribution from such a reaction, if any, is not large and it is not predominating.

The yield of  $At^{211}$  from lead foils of different thickness is given in Fig. 2. The figure shows that the yield of  $At^{211}$  does not change practically for thicknesses ranging from 0.3 to 0.6 mm. Below the thickness of 0.3 mm the yield decreases monotonously and with the target 0.03 mm thick it is half the yield on the plato.

### **Discussion of the Results**

The observed quantatity of astatine cannot be formed from spallation of possible uranium and thorium contaminations and secondary reactions of Bi contaminations. The evaluations show that only 0.1 of the yield of  $At^{211}$  and  $At^{210}$  can be explained by spallation of possible contaminations of the above elements. With protons of energy higher than 120 MeV this fraction becomes still smaller.

Thus, the observed activity of At is entirely due to the secondary reaction of Li-capture.

The dependences of total astatine yields from lead and the analogous reactions of iodine-formation from tin<sup>9/</sup> on the energy of incident protons (see Fig. 3) have been compared. As is seen, dependences are similar. At 660 MeV the yields of astatine and iodine coincide.

The total yield of astatine (0.2 microbarn) observed in our experiments at proton energy of 340 MeV agrees satisfactorily with the yield of the analogous reaction of lead formation from Au (0.36 microbarn) at proton energy of 340 MeV/11/.

The estimates of energy spectra of Li fragments and their formation cross section from lead bombarded with high energy protons have been made as described in  $^{10/}$ .

It was necessary to choose such a form of Li fragments so as to obtain the relative yields of At isotopes when the excitation functions of Li-capture reactions and the energy losses of Li-fragments for ionisation are known. When the spectrum was chosen, the cross section of Li was calculated. Basing on  $^{/5/}$  we have chosen Li spectrum as

$$P(E)dE = \frac{E-V}{C^2} \cdot e^{-\frac{E-V}{C}} dE$$

selecting corresponding parameter values of V and  $\tilde{c}$ . It was assumed that the energy spectra of  $Li^6$ ,  $Li^7$  and  $Li^8$  isotopes are the same<sup>/5/</sup>. Excitation functions for reactions of Li-capture in lead isotopes were calculated by Jackson formulas<sup>/20/</sup> separately for  $Li^6$ ,  $Li^7$  and  $Li^8$ . The cross section of Li-capture in lead was calculated by the Babikov formula<sup>/21/</sup>.

Fig. 4 displays the calculated excitation functions for basic formation reactions of  $At^{211}$ ,  $At^{210}$  and  $At^{207}$  for  $Li^6$  — and  $Li^7$  — capture in different lead isotopes. 25 from 43 possible reactions have been considered. In each case the prevalence of the corresponding lead isotope and the relative yield of  $Li^6$ ,  $Li^7$  and  $LI^8$  in spallation were taken into account. The relative yield of  $Li^6$ :  $Li^7$ :  $Li^8$ : fragments<sup>/11/</sup> from lead has been taken as well as for Au equal to 0.55: 0.41: 0.043. Range energy relations of Li fragments in lead were calculated by known formulas<sup>/22/</sup>.

Relative calculation yields  $\frac{At^{210}}{At^{211}}$  and  $\frac{At^{207}}{At^{211}}$  and also the formation cross section of Li fragments with energies>30 MeV at an energy of 660 MeV for different parameter values of V and  $\mathcal{T}$  are enlisted in Table 2. As is seen from Table 2, the critical value which determines the energy spectrum is the

\* We do not imply any physical sense in the employed formula of the energy spectrum. One can take the dependences of the type  $\rho(E)dE = \frac{1}{En}dE$  where  $n \simeq 2$ . However, the chosen dependence approximates somewhat better all the known experimental data.

V	Т	<u>At</u> 210 At211	<u>Al</u> 207 At211	с <sub>и</sub> , тв	
	11.5	1.67	0.9	6.0	
6	10.5	- 1.63	0.79	6.1	
	9.5	1.57	0.63	6.7	
•	11.5	1.7 <sup>*****</sup>	0.86	5.7	
10	10.5	1.65	0.8	6 <b>.0</b>	
	9.5	1.57	0.63	6.7	
	6.5	1.39	0.28	9.4	
15	5,5	1.27	0.15	11.5	
and the second	4.5	1.16	0.09	14.5	

Table 2.

Li-fragments with an energy>54 MeV from Au turned out to be four times greater than in our case at an energy of 340 MeV (from Au  $G_{L} = 1$  millibarn, from Pb  $G_{L} = 0.27$  millibarn).

The approximate constancy of the relative yield 660 MeV and 120 MeV (see Table 1) points out that the spectrum of over Coulomb Li fragments either does not depend on proton energy or changes negligibly.

Aiming to check up the obtained spectrum of Li fragments, we calculated the probability of At<sup>211</sup> as a function of lead foil thickness and compared it with the dependence observed experimentally. In order to perform calculations it is necessary to know the effective Li range in foils of different thickness. These ranges were evaluated on the basis of the angular distribution of fast Li fragments quoted in<sup>6</sup>/. The probability of At<sup>211</sup> production in a lead layer with the thickness 'd' was calculated as the difference between the probability of  $W_{\bullet}$  production of this isotope by a fragment of the given energy in a lead layer of infinite thickness and as the difference of the probability W of the in-

dicated product formation by a fragment with an energy which remains on passing a lead layer with the thickness 'd'. Here we have

$$W_{o} = \sum_{i=1}^{H} N_{o} G_{i} \Delta l_{i}$$
<sup>(2)</sup>

where  $N_0$  - is the number of lead nuclei in  $1 \text{ cm}^3$  and  $G_i$  is the cross section of Li capture which leads to the formation of  $At^{211}$ , and  $\Delta \ell_i$  is the ionisation range of a fragment in lead at energies ( $E_i + \Delta E_i$ ;  $E_i$ ). For each thickness of 'd' numerical integration of  $At^{211}$  formation probability was performed along the energy spectrum of Li.

The calculated dependence of  $At^{211}$  formation probability is given in Fig. 2 in the form of a curve. It was matched with experimental points at the thicknesses equal to 0.24 mm and even more. As is shown by figure 2 the curve satisfactorily describes the decay of experimental points with small thicknesses of lead foils. The suggestion made in the process of calculation ( on the spectra identity of different Li isotopes and also on the magnitude of the ratio  $Li^6$ :  $Li^7: Li^8$ .) and possible deflections of the calculation excitation functions from the real ones decreases the reliability of conclusion arising as a matter of spectrum evaluation and the dependence of the yield on foil thickness. Consequently, these conclusions should be considered only qualitatively.

The dependence of  $At^{211}$  formation probability on the lead foil thickness observed in our experiment permits to evaluate independently the cross section of over Coulomb' Li fragments. The cross section (note it as  $G_{Li}^{P}$ ) is found from the equation

$$B = N_{o} \cdot G_{Li}^{\rho} \cdot G_{At} \cdot \Delta t$$

(3)

In this formula  $\mathcal{B}$  - the yield of astatine for the given proton energy,  $\mathcal{G}_{At}^{\mathcal{L}_i}$  is the reaction cross section of Li-capture in lead isotopes averaged by energies which leads to astatine formation. (In this case the isotope At<sup>211</sup>). The values of  $\mathcal{B}$  and  $\Delta \ell$  are obtained directly from Fig. 2.  $\mathcal{B}$  is the ordinate of the curve in the saturation region and  $\Delta \ell$  corresponds, roughtly speaking, to the half of the lead foil thickness in which the probability of At<sup>211</sup> production starts to deflect from saturation. The value of  $\mathcal{G}_{At}^{\mathcal{L}_i}$  was calculated from excitation functions for the corresponding reactions of At<sup>211</sup> formation, the prevalence of lead isotopes and different Li yields in spallation being taken into account. The value  $\mathcal{G}_{At}^{\mathcal{L}_i}$  is equal to 0.1 barn. Substituting the above calculated values to formula (3) we obtain  $\mathcal{G}_{\mathcal{L}_i}^{\mathcal{L}_i} = 3-4$  millibarn. This value agrees satisfactorily with the cross section of over Coulomb Li fragments calculated from energy spectra.

Here are some remarks on the mechanism of over Coulomb barrier fragments formation. Despite the fact that several authors have explained some peculiarities of the process of fragmentation from the point of view of the statistical model/23,24/ this model turns out to be unacceptable to explain the ori-

gin of over Coulomb barrier fragments. The statistical model accounts for the most number of fragments having energies close to the energy of the Coulomb repulsion. However, a considerable part of over Coulomb fragments cannot be explained by the statistical model. It can be seen, for instance, from  $^{4/4}$  in which the energy spectra of fragments have been studied with the aid of emulsions. The authors of the above paper speak about a partial success of the evaporation theory and indicate the necessity to involve a new mechanism. The unacceptability of the statistical model for explaining the energy spectra of over Coulomb fragments is shown in our paper also. If the parameter  $\mathcal{C} = 10.5 - 11.5$  MeV implies physical sense of nuclear temperature as it is required by the evaporation theory this leads to absurd results since nuclear excitation energy turns out to be several times higher than the energy of incident particles. On the other hand the use of the parameter values  $\vee$  and  $\mathcal{C}$  in formula (1) which are reasonable from the point of view of the evaporation theory ( $\vee = 15$  MeV and  $\mathcal{C} = 4.5 - 5.5$  MeV) provides, as it is seen from Table 2, the magnitude of the ratio  $\frac{At^{207}}{At^{21}}$  considerably less than the experimental one.

The formation of over Coulomb barrier fragments is difficult to explain from the point of view of the statistical model by possible 'local' overheating of the nucleus caused, for instance, by the process of production and reabsorption of pions<sup>25,26/</sup>. As is shown in Fig. 1 the production of over Coulomb barrier fragments occurs also at energies of incident particles which lay considerably lower than the mesoproduction threshold.

The ejection of over Coulomb barrier fragments evidently occurs essentially earlier before the nuclear heating takes place when, in general, the statistical treatment is unacceptable. It is proved, for example, by the relation between the yield of high energy fragments and by the number of cascade particles in splittings<sup>/4/</sup> and also by the forward direction of emerging over Coulomb fragments marked nearly in all the papers on observation of fragments in photoemulsions. The standpoint according to which the formation of over Coulomb fragments is treated as a result of direct acts of multiparticle interactions of the incident particle as well as of the cascade nucleons with nuclear nucleons/27/ seems to be more promising. One can imagine such interactions suggesting that sometimes nucleons can approach each other in the nuclear matter for a short time by fluctuations to distances shorter than they do it in a usual nucleus. Under such conditions the incident particle can interact with a fluctuation group of nucleons as with a whole and can transfer it a considerable part of its own energy. However, the model of the fluctuation compression of nuclear matter as it is worked up for the explanation of the emergence of high energy deuterons<sup>28/</sup> is not acceptable in our case. It gives a very small probability of over Coulomb Li fragments formation and does not account for different dependences of the yields of these fragments in bombardment with protons and **&-** particles.

It should be noted that acts of multiparticle interactions are responsible for over Coulomb barrier fragments, the inverse events should occur in reactions with multiply charged ions, when the energy of the incident ion is carried away by single nucleons. Apparently, events observed in  $^{28/}$ , when the whole excitation energy (  $\sim$  60 MeV) is carried away by two nucleons, points out the possibility of arising of such inverse multiparticle interactions.

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The dependence of the yield of the isotope  $At^{211}$  (Gat20) on the energy of incident  $\checkmark$ -particles ( $\checkmark$ ), deuterons (d) and protons (P).





The dependence of the total formation cross section of a statine isotopes from lead and iodine isotopes from tin on proton energy.



Excitation functions for fundamental reactions of  $At^{211}$ ,  $At^{210}$  and  $At^{207}$  formation in  $Li^6$  – and  $Li^7$  –capture in different lead isotopes.



