The Astatine Isotopes Produced in He^3 Bombardment of Bismuth

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Bi를 He^3로 照射하였을때 生産되는 At 同位元素

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Abstract

The astatine isotopes produced in He^3 bombardment of bismuth were analysed with alpha ionization chamber and 100 channel pulse height analyser. The main product was At^{209} formed by (He^3, 3n) reaction. Four other isotopes were also identified. Semi-quantitative calculation of relative yield of each isotope was attempted.

要 約

Bi^{209}를 He^3로 照射했을때 생긴 At 同位元素를 Alpha Ionization Chamber 및 100 Channel 波高分析器로 調査하였다. 主生成物은(He^3, 3n) 反應으로 생기는 At^{209}였고, 그外에도 4種의 At 同位元素를 얻었다. 各 同位元素의 相對的인 收量을 半定量的으로 計算하였다.

Introduction

The production of astatine isotopes by the alpha particle bombardment on bismuth was reported by Kelly and Segre[1]. Depending on the bombarding energy of alpha particles, different astatine isotopes were preferentially produced. With the alpha particles in the energy range of 29–30 MeV, the cross section of the reaction $Bi^{209}(\alpha, 2n)At^{211}$ reaches its maximum and$(\alpha, 3n)$ reaction just begins.

A number of new neutron deficient astatine isotopes have been identified later in the mass number range 200 to 209 following the irradiation of bismuth with high energy helium ions by Barton et al.[2]. They detected $(\alpha, 4n)$ and $(\alpha, 5n)$ reaction at about 55 MeV.

In the present study, bismuth targets of thickness 3 to 4 mg per cm^2 were bombarded in the 30 MeV external He^3 beam of the 60 inch cyclotron of the University of Birmingham, England. The reaction products of astatine were then separated and analysed using alpha ionization chamber and 100 channel pulse height analyser. The main product was At^{209} formed by (He^3, 3n) reaction. In addition to this, the astatine isotopes, At^{211}, At^{210}, At^{208}, and At^{207} were identified.

In identifying the isotopes, the pulse analysis of their electron capture decay products were also used and in the case of At^{207} the milking experiment was carried out.

Preparation of Material and Chemical Procedure

The bismuth targets that were bombarded were produced by evaporating bismuth from a tungsten boat in a high vacuum upon aluminium discs of 60 micron thickness. Bismuth targets prepared in this way were of uniform thickness of about 3 to 4 mg per cm^2.[3]. These samples were then clamped in a water-cooled target probe and bombarded in the external beam of He^3. The

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maximum energy of 30 MeV was used in all bombardments with beam current of 1.5μ amp. for 20 min. periods.

After the bombardment, the bismuth target was removed from the probe and dissolved with 2-3 drops of concentrated nitric acid. This target solution contains isotopes of polonium, bismuth, lead and thallium besides the mixture of astatine isotopes. To obtain a pure astatine fraction without carrier, the dissolved target solution made to 8N HCl was extracted with diisopropyl ether (DIPE).[4]

A fraction of the DIPE solution was then evaporated on platinum or silver discs for alpha-pulse analysis.

For the identification of At^{207} through polonium daughter grown by K-capture decay, the daughter products were removed by adding two-hundredth volume of tributyl phosphate to the DIPE solution and extracting into 2M nitric acid-4M hydrochloric acid solution. With about one minute contact, all the daughter elements leave the organic phase while the astatine remains in the organic layer.

The daughters, mainly polonium, were then coprecipitated with bismuth hydroxide and dried on a filter paper and counted with an end-window Geiger counter. This "milking" process[1] was repeated on the parent solution at equal time interval.

Although other daughters formed during the interval were also coprecipitated with the bismuth hydroxide, the half-live of parents and daughteres exclude the possibility of complication by the contamination to the result. (See Fig. 1)

To obtain a thinner astatine sample than the one from the above chemical operation, double evaporation procedure was tried. The target was melted by heating in a glass tube and the astatine evaporated was caught by aluminium or platinum disc attached on a cold with ice. This was repeated twice to give a pure astatine source. The source prepared in this manner was no better than the one from solvent extraction.

Apparatus

The electrode arrangement of the grid chamber used in this work was essentially the same as the one by Harvey et al.[5]. It consisted of two plane parallel electrodes 6.5cm apart and the grid and a guard ring in between. The lower 10.2cm^2 brass plate had a 3cm diameter hole at the center for the source disc. Alpha particles were collimated by a ring 0.3cm high with a diameter of 3cm which was concentric with the source disc. 5.5cm away from this source plate was the grid consisting of a frame 10.2cm^2, strung with 0.005 inch copper wires, 0.9mm apart. A guard ring was used to improve the uniformity of the field lines between the collector and source. This

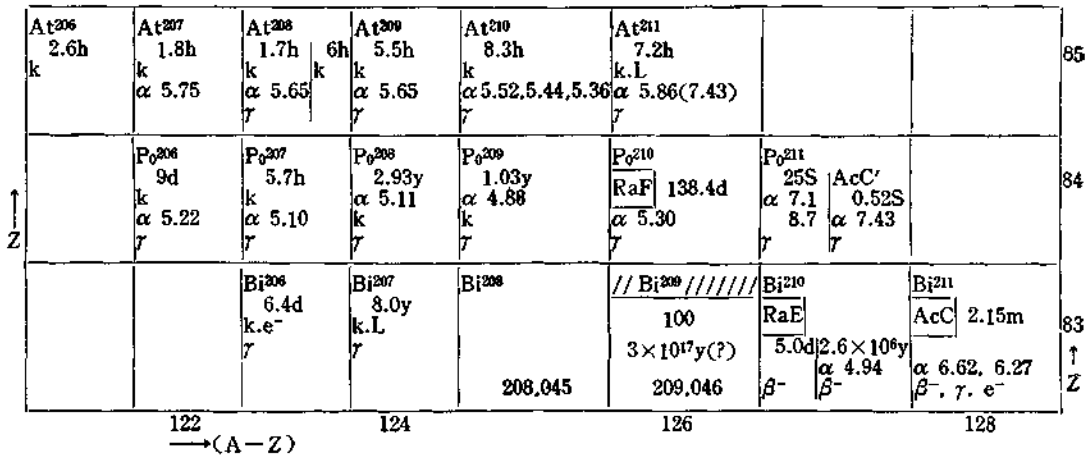

Fig. 1 Isotope chart of Bi-At region

was placed 3.5cm away from the source. The upper plate was the collector electrode made of 10.2cm diameter brass disc. Referred to the collector, which was at ground potential, the grid was at −800 volts, the guard ring was at −1.4k volts, and the source plate at −2.0k volts.

In the chamber, a six position turntable was provided with an external control. The radioactive sources used in this experiment were placed on the turntable and each source could be brought into the counting position by the external control. Therefore all the sources were counted under identical conditions of geometry and gas filling.

The chamber was filled to a pressure of 11 pounds per square inch above the atmospheric pressure with argon-methane mixture(90-10%), after evacuating the chamber to less than 0.03mm Hg, which usually took about an hour of pumping with the ordinary rotary pump. The small variation in pumping period and the pressure of filling gas did not affect the result of pulse analysis.

Fig. 2 shows a block diagram of the electronic equipment. The head amplifier was mounted directly on the chamber so that the high signal to noise ratio could be obtained. The time constants and attenuation settings of the main amplifier, were chosen experimentally to give optimum conditions for the pulse analysis. The 100 channel pulse analyser used in this work was model H. S. 100 designed and manufactured by Marshall of Cambridge Electronics Ltd. This also had a time integrater, which gave a live time and a recorder to record the number of pulses in each channel.

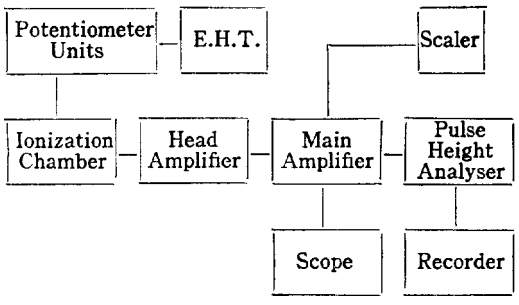

Fig. 2 Block diagram of electronic equipment

Calibration

Calibration of pulse height vs. energy was carried out by the use of standard samples containing alpha emitters of known energy[6]. The standards used in this work were Th^{232} and its daughters, Po^{210} and thorium active deposit from the strong radiothorium source provided in the laboratory. The thorium active deposit ThB, which gave 6.05 MeV ThC and 8.78 MeV ThC' peaks, was deposited on the other two standards. The Th^{232} standard was prepared by evaporating a solution of thorium nitrate in absolute ethanol on a platinum disc. This standard had five well distinguished peaks and a peak from ThC. (Fig. 3). Po^{210} standard was prepared by the usual electrodeposition method[7].

Because of the instrument drift in calibration, it was necessary to use an internal standard. The drift, however,, was not serious during 6 to 8 hours of continuous operation, although the calcium furnace provided for the removal of traces of oxygen and other electron-capturing impurities from the gas was not used. Average of about 1 channel drift in 3 hours was found. Once the calibration was made accurately, one internal energy standard was sufficient to test the possibility of drift.

Results and Discussion

In determining the relative abundance of alpha

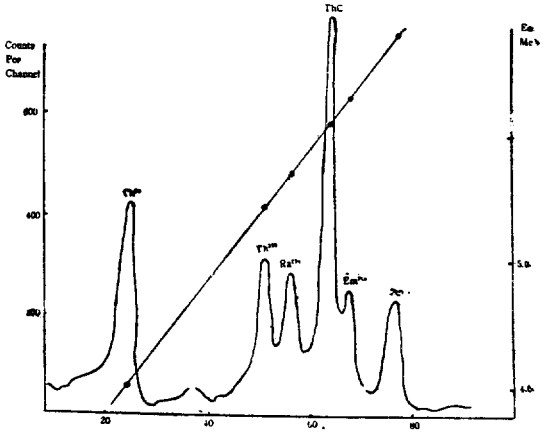

Fig. 3 Th Standard. Bias 15V

activiy at the vaious peaks, the counts in the channels involved at each peak were simply added up. The comparison made in this manner was more reliable than comparison of peak heights because of the possible variation of straggling parameter [6] from peak to peak. When two peaks overlapped, the intermediate region had to be approximately divided by drawing the overlapping high and low energy tails to correspond to the shapes of the non-overlapping parts. From this relative abundance of alpha activity, the activity at the end of bombardment was extra-polated back and the saturation activity was then obtained by dividing by $(1-e^{-\lambda t})$. This is the rate of formation of the isotopes by definition. (λ = decay constant, and t = bombardment time.)

In order to obtain the saturation activities of At^{210}, At^{209} and At^{208} from the corresponding resolved polonium daughter activities, the general equation representing the growth of a radioactive daughter(subscript 2) formed in the decay of the parent substance(subscript 1) was used. Since the pure astatine was extracted by DIPE after the bombardment, it could be assumed that no polonium was present initially, The symbols have their usual connotations[8].

$$\lambda_2 N_2 = \frac{\lambda_1\lambda_2}{\lambda_2-\lambda_1} N_1^0(e^{-\lambda_1 t'} - e^{-\lambda_2 t'}) \qquad (1)$$

The left-hand side is the activity of the daughter at any time measured from the end of bombardment. The right-hand side represents the growth of daughter from parent. t' is the time measured from the end of bombardment.

Since $\lambda_1 \gg \lambda_2$ in all cases, the daughter activity can be represented simply by,

$$\lambda_2 N_2 = N_1^0 \lambda_2 e^{-\lambda_2 t'} \qquad (2)$$

for $t' \gg T_1$ where T_1 is the half-life of the parent,

$$\lambda_1 N_1^0 = \frac{\lambda_1}{\lambda_2} \cdot \lambda_2 N_2 e^{\lambda_2 t'} \qquad (3)$$

$$\therefore \quad \lambda_1 N_1^0 = \frac{\lambda_1}{\lambda_2} A_2^0, \qquad (4)$$

Where A_2^0 is the daughter activity extrapholated back to the end of bombardment.

Putting, $\lambda_1 N_1^0 = R(1 - e^{-\lambda_1 t})$

we obtain an expression for the saturation activity of parent.

$$R = \frac{\lambda_1 A_2^0}{\lambda_2(1-e^{-\lambda_1 t})} \qquad (5)$$

The initial astatine alpha pulse analysis showed that the strong activities of 5.65 MeV group have completely obscured 5.75 MeV group of At^{207} and 5.86 MeV group of At^{211} partly. Therefore, the additional collimator of 1/32 inch thick with many holes of 1/16 to 1/8 inch size was used in an attempt to improve resolution[9], and slightly improved spectrum was obtained, From this assumed 5.75 MeV peak, and 7.43 MeV peak, and the relative abundance of At^{207} and At^{211} were calculated. The presence of At^{207} in the bombarded sample was later confirmed by milking experiment as shown in Fig 4. This shows 1.8 hour half-life of At^{207} and also the decay of the first milking sample gives 5.7 hour Po^{207}.

The decay of 5.65 MeV peak and 7.43 MeV peak was followed separately by successive pulse analysis.

After leaving the sample for 6 to 7 days, the final pulse analysis was carried out. This time, all the astatine activities had been decayed and only their long-lived daughter Po activities remained.(Fig.5)

The activities were rather low because of their long half-lives, but the relative yields of their parents, At^{208}, At^{209} and At^{210} were calculated based on this relative abundance of polonium activities. The branching ratio of each isotope

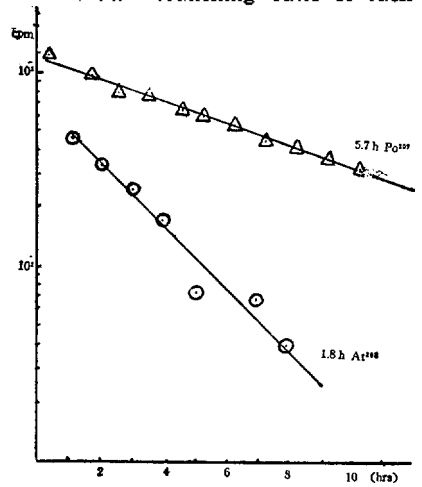

Fig. 4 Decay curves from milking experiment

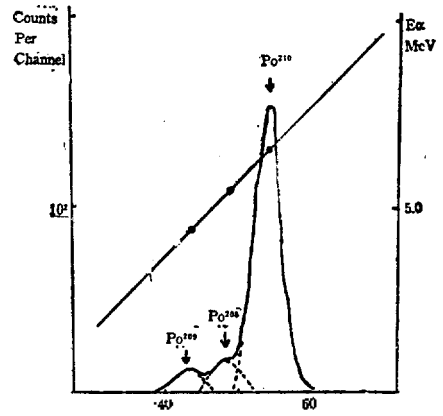

Fig. 5 Pulse analysis of Po. Bias 20V

used in the calculation and the results obtained are tabulated in Table 1.

The straggling parameter measured in this work showed about 3 times greater than that of other authors who used the same apparatus but differnt electronic equipment(100 KeV compared to 30 KeV).

The improvement in electronics for better resolution is essential for the accurate work with the apparatus used in this experiment.

The yield is the rate at which new activity is formed, under specified bombardment conditions of particle energy and current, etc.[10]. If we assume uniform bombardment and constant current, then

$$R=\frac{\lambda N}{(1-e^{-\lambda t})} \qquad (6)$$

where

λN = activity at any time, t,

Table 1 Results and other data

Product	Reaction involved	Relative yield(%)	Half-life	Energy MeV	Branching ratio(%)
At^{207}	(He^3, 5n)	<0.5	1.8hr. (Po^{207}= 5.7hr.)	5.75	At^{207} α=10 E.C.=90
At^{208}	(He^3,4n)	3	(1.7 hr.) (6.hr)	5.65 (Po^{208}= 5.11)	At^{207} E.C=100 / (Po^{208} α=100)
At^{209}	(He^3,3n)	92	5.5hr	5.65 (Po^{209} = 4.88)	At^{209} E.C=95 α=5 / (Po^{209} α=90) E.C.=20)
At^{210}	(He^3,2n)	4	(8.3hr.)	(Po^{210}= 5.30)	At^{210} α=99.8 E.C=0.2 / (Po^{210} α=100
At^{211}	(He^3, n)	<0.5	7.2hr.	5.86 (Po^{211}= 7.43)	At^{211} α=41 E.C.=59

R = constant rate of production,

If, t = bombardment time,

then, λN is the activity at the end of bombardment. In all bombardments, target thickness of average 3.5 mg/cm² was used and the target area of 2.25 cm² was irradiated by the average of 1.5 μ amp. for 20 minutes. These conditions were not changed from one bombardment to another.

Some typical calculations of relative yield are as follows.

At^{221}

Alpha activity at the end of bombardment = 224 cpm(corrected for the counter efficiency)

For total DIPE solution,=16.777 cpm,(corrected for α-branching.)

Saturation activity,=0.61×10^6 cpm,(from eq. [6]).

Yield. $=0.41\times10^6$d.p.m./(μa.)

At^{210}(Po^{210})

The total corrected activity of Po^{210} at the end of bombardment,

$A_2^0 = 55614$ cph.

$\lambda^1=\frac{0.693}{8.3}$, $\lambda^2=\frac{0.693}{3321.6}$

∴ From equation(5),

$R=809.3\times10^6$ cph$=13.5\times10^6$ cpm.

correcting for alpha branching,

$$13.5\times10^6\times\frac{100}{99.8}=13.5\times10^6 \text{ cpm.}$$

yield$=8.9\times10^6$ dpm/(μa.).

The unit of yield is atoms disintegrating per minute for 20 minutes bombardment of 3.5 mg/cm² thick Bi of 2.25 cm² by 30 MeV He^3 of one μa. current beam.

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