SEARCH FOR SECONDARY DEUTERIUM AND TRITIUM CAPTURE REACTIONS

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The yields of heavy arsenic isotopes produced by bombarding germanium with 120, 300, 480, and 660-MeV protons are determined by a radiochemical method. With increasing proton energy all yields decrease monotonically; for the As^{74} nucleus the yield is between 3.4 and 1.0 mb, for As^{76} between 1.0 and 0.38 mb, and for As^{77} and As^{78} between 0.13 and 0.035 mb. The mechanism for the production of As^{74} and As^{76} is apparently the primary (p, xn) reaction. The As^{78} isotope is apparently produced as a result of the capture of superbarrier tritium nuclei. The origin of the As^{77} isotope is more complex. At low proton energies (120 and 300 MeV) it is produced mainly as a result of the secondary capture of deuterium and tritium. At higher proton energies most of this isotope is apparently obtained as a result of secondary reactions of α-particle capture.

INTRODUCTION

UP to now it has been possible to observe in radiochemical investigations of "secondary" reactions[1-9] capture reactions of superbarrier α particles,[1-7] lithium nuclei,[1-5,8,9] and sometimes beryllium nuclei.[2] It would be interesting to attempt to extend the range of observable reactions, in particular to study the features of the capture reactions of superbarrier deuterium and tritium nuclei. Experiment shows that high-energy deuterium and tritium nuclei are emitted quite often in the bombardment of compound nuclei by ultrafast protons,[10,11] and consequently possibilities exist for secondary capture reactions of such fragments.

Just as in the investigation of secondary reactions, the effects expected here are small; for this reason such experiments require the irradiation of extremely pure materials and particular care in the chemical separation and purification operations. Another difficulty inherent in such experiments is the fact that the primary (p, xn) reactions (x = 1, 2 is the number of emitted neutrons), whose yields exceed the cross sections of the sought products by a factor of 100 or more, interfere to a large extent with the observation of the capture reactions of singly-charged fragments. This explains the specific requirements regarding the choice of the irradiated element, and the method of registration of the reaction products.

1. EXPERIMENTAL METHOD

Because of its high purity, we chose as the irradiated element germanium used in the production of semiconductors. The element sought in this case was arsenic. The interfering (p, xn) reactions yield neutron-deficient arsenic isotopes (from As^{70} to As^{74}), the latter being produced from all the stable isotopes of germanium. At the same time one can expect the production of heavy arsenic isotopes (As^{76-78}) chiefly from the secondary capture reactions of deuterium and tritium by the Ge^{74} and Ge^{76} isotopes. The natural abundance of Ge^{74} and Ge^{76} amounts to 36.7 and 7.7 percent respectively, and the contributions of secondary reactions can still be appreciable.

Samples of powdered germanium weighing 0.2—0.5 g were packed in aluminum foil, and irradiated in the internal proton beam of the synchrocyclotron of the Joint Institute for Nuclear Research. The energy of the bombarding protons was changed by setting up the samples on proton orbits of different radii. The time of irradiation usually amounted to about 1 hour. After irradiation, the germanium samples were dissolved in aqua regia into which 10 mg As^{V} and hold-back carriers were previously introduced. The solution was transferred into a hydrochloric solution by evaporation with concentrated hydrochloric acid. In the course of this a considerable part of the germanium was distilled in the form of $GeCl_4$. The remaining germanium

was extracted 2—3 times with benzene. Then HBr was added to the solution to reduce the As^{V} to As^{III} and the arsenic was extracted with benzene. [12,13] The arsenic was extracted from the benzene layer with the aid of a hydrazene solution, and was precipitated after oxidation in the form of a sulfide. The final stage of purification was the obtaining of metallic arsenic. [14] Metallic arsenic deposited on filter paper was needed for the measurements. The entire chemical separation took 3—4 hours, and the chemical yield amounted as a rule to 30—40 percent.

The arsenic samples were measured with the aid of an end-window beta counter with an electromagnet [15] in such a way that only the electron component of the emitted radiation was directed into the counter. In order to observe electrons with different energies, the measurements were conducted with various values of the electromagnet current. For each chosen value of the current we followed the decrease of the activity with time. It was thus possible to observe the half-lives of the electrons of each of the isotopes of interest to us: As^{74}, As^{76}, As^{77}, and As^{78}. The method chosen allows, along with the registration of the electrons from the decay of the heavy isotopes, also partial registration of the conversion electrons emitted in the gamma transitions in the neutron-deficient isotopes. In our opinion, the error introduced here is small, since in the light arsenic isotopes the high-energy conversion electrons (we detected electrons with energies of 0.4 MeV and higher) are relatively weakly pronounced, and can in addition be easily separated by their half-lives. The same detector was used to measure the Na^{24} samples from the aluminum foil serving as the proton-beam monitor. The monitor samples were prepared by separating a definite portion of an aluminum solution in hydrochloric acid, and drying it on blotting paper.

The decay curves of the arsenic samples for all values of the electromagnet current included the following lifetimes: ~ 1.5 hour, 24—27 hours, and 17—19 days. The first and last lifetimes were investigated during three half-lives, while the activity with the lifetime of 26 hours was investigated during 5—6 half-lives. For small values of the current (0.5 and 1.0 A) we observed a small contribution of an activity with a half-life of 38—42 hours. The determination of the contribution of the latter on the background of the much stronger 24—27 hour activity was accomplished with the aid of a special mathematical method of transforming the results of the measurements [1] on the portions of the decay curves of interest to us, which made it possible to display better the required component. The mathematical transformation was applied to the decay curves obtained with small currents (0—2.0 A). The recalculated activities were unambiguously identified [16] as As^{78}, As^{76}, As^{74}, and As^{77} respectively. The identification of the observed activities was confirmed by special measurements of the conversion-electron spectra of the arsenic samples [2] carried out immediately after the chemical separation. Clearly resolved peaks with energies of 555 keV (As^{76}), 580 keV (576 keV Ge^{69} + 596 keV As^{74}) and 612 keV (As^{78}) were observed in the energy range of 500—650 keV. There are indications that a peak exists at 525 keV (As^{77}).

The yields of the above arsenic isotopes were calculated as in [17] with allowance for the time of irradiation and the decay of the sample after the irradiation, and also with allowance for the chemical yields. The initial activities for the calculation of the yield of the separate components were taken to be the areas under the curves plotted from the change of the number of counts of the detector as a function of the current in the electromagnet. These curves were obtained by separating into the component half-lives the general curves of the decrease of the number of counts registered by the detector at a given value of the current. The current in the electromagnet could be changed from 0 to 3 or 5 A. In this range of values of the current the entire electron emission of As^{77}, As^{74}, and Na^{24} (end-point energy ≤ 1.4 MeV) was directed by the electromagnet into the detector, and the determinations of the yields of the enumerated arsenic isotopes were carried out with complete efficiency. The activities of the As^{76} and As^{78} nuclei were not fully registered by our measuring device, since in the spectrum of these isotopes there is a considerable fraction of hard electrons

[1] The method consists of the following: assume we have a decay curve

$$A(t) = A_k \exp(-\lambda_k t) + A_n \exp(-\lambda_n t), \quad (1)$$

where A_k and A_n are the initial activities of the components k and n which are to be determined; λ_k and λ_n are the decay constants. We multiply (1) by $\exp(\lambda_k t)$; then

$$A(t) \exp(\lambda_k t) = A_k + A_n \exp[(\lambda_k - \lambda_n t)]. \quad (2)$$

Setting,

$$A(t) \exp(\lambda_k t) = y, \quad (3)$$

$$\exp[(\lambda_k - \lambda_n) t] = x, \quad (4)$$

we obtain on the xy plane the equation of the straight line $y = A_k + A_n x$.

[2] We express our gratitude to Zh. Zhelev of the Joint Institute for Nuclear Research, who carried out the measurements of the conversion-electron spectra of arsenic on the beta spectrometer.

Proton energy, MeV	Production cross sections, mb							
	As^{74}		As^{76}		As^{77}		As^{78}	
	Experimental	Theoretical	Experimental	Theoretical	Experimental	Theoretical	Experimental	Theoretical
120	3.4±0.8	4.8 a, d) 7.4 e)	1.0±0.15	0.9 a, d) 1.1 e) 0.9 b) 0.5 c)	~0.1	—	0.13±0.045	— 0.003 g)
300	2.8±0.9	1.7 d, a) 3.0 e)	0.83±0.2	0. d, a) 0.5 e)	~0.08	<0.035— —0.038 f)	0.09±50.021	—<0.015 h)
480	1.6±0.4	0.8 d)	0.52±0.13	0.2 d)	~0.07	—	0.073±0.016	—
660	1.0±0.6	0.6 d)	0.38±0.13	0.1 d)	~0.04	—	0.035±0.007	—
Possible production reaction	$\frac{Ge^{74}(p,n)}{Ge^{76}(p,\ 3n)}$		$\underline{Ge^{76}(p,\ n)}$		$Ge^{76}[p(d,n)]$ $Ge^{76}[p(H^3,\ 2n)]$ $Ge^{74}[p(\alpha,\ p)]$ $Ge^{76}[p(\alpha,p2n)]$		$Ge^{76}[p(H^3,n)]$ $Ge^{76}[p(\alpha,pn)]$	

The estimates of the expected yields are obtained from:

a) the yield of the (p, n) reaction on Ga^{69} at a proton energy of 100 MeV,[22]

b) the yield of the (p, n) reaction on Zn^{66} at a proton energy of 100 MeV,[22]

c) the yield of the (p, n) reaction on Cu^{63} at a proton energy of 100 MeV,[23]

d) the yield of the (p, xn) reaction on Ga^{69} at energies of 0.5, 1.5, and 2.9 BeV,[24]

e) the yield of the (p, xn) reaction on Y^{89} at energies of 100-240 MeV,[25]

f) the sums of the estimated yields of the secondary [p(α, p)] and [p(α, p2n)] reactions, amounting to 0.028 and 0.007-0.010 mb respectively. In determining the yield of the [p(α, p)] reaction we have made use of the cross section of the [p(α, n)] reaction on copper at 340 MeV,[1] which is 0.01 mb, and also of the ratio of the integral yields of the (α, p) and (α, n) reactions on Zn^{64}[20] which is 2.4. In estimating the yield of the [p(α, p2n)] reaction it has been assumed that the fraction of the [p(α,3n)] reaction amounts to no more than 1/3 of the cross section of the [p(α, n)] reaction on copper, and the ratio of the integral yields of the (α, p2n) and (α, 3n) reactions on Zn^{64} is 10,[20]

g) the yield of the [p(α, pn)] reaction using the 0.006-mb cross section of the [p(α, 2n)] reaction on copper,[1] and the ratio of the integral yields of the (α, pn) and (α, 2n) reactions for a Ge^{70} target which is 2,[26]

h) the limit of the possible contribution of the [p(α, pn)] reaction [obtained by using the ratio of the integral yields of the (α, pn) and (α, 2n) reactions which is 10 for Zn^{64}[20]].

(3 and 4.1 MeV) which were not directed into the detector at the maximum available electromagnet current. For this reason the initial activities of As^{76} and As^{78} had to be multiplied by the coefficients k, equal to 1.8 and 2.3 respectively. The correction coefficients were estimated from the mean energies of the electrons emitted by the given isotope. In calculating the As^{77} yield, corrections were introduced for self-absorption and the absorption of beta particles in the outer cover of the sample, the counter, and the layer of air (k = 1.8).

2. EXPERIMENTAL RESULTS

The identified heavy isotopes of arsenic, and their production cross sections are presented in the table. The mean of two, and sometimes of three determinations is given in each case. In most of the experiments the results of individual measurements agreed with each other within ±25 percent. In only one instance (the yield of As^{76} at 660 MeV) did they differ by ±40 percent. Nonetheless, for a number of reasons we recommend that the absolute cross sections presented be considered correct within a factor of 2. The table also indicates the possible reactions in which the enumerated arsenic isotopes are produced, and gives their contributions as expected from the abundances of the initial nuclei. Among the possible reactions we include the primary (p, xn) reactions on the bombarding protons (underlined) and the secondary reactions of α-particle, deuterium, and tritium capture accompanied by nucleon emission. In view of their small yield,[18-21] the secondary radiative-capture reactions of the above particles, as well as the reactions of capture of He^3 nuclei, are not taken into account.

As can be seen from the table, the greatest yield is that of the As^{74} isotope. The production cross section of As^{76} is smaller by about a factor of 3, and the yields of As^{77} and As^{78} are almost the same, and by an order of magnitude smaller than the yield of As^{76}. With decreasing proton energy the production cross sections of all observed nuclei decrease equally.

Comparing the experimental and the expected yields of As^{74}, as well as of As^{76}, we see that they are close to one another and exhibit approximately the same dependence on the proton energy. Particularly good agreement is obtained for As^{77} where the deviation between the expected and observed cross sections never exceeds a factor of 2. Such a deviation is quite acceptable and can be due, on the one hand to errors in the estimates of the expected yields depending on the choice of target, and on the other hand to errors in the determination of the absolute cross sections in our experiments. In the case of As^{76} the deviation observed at low proton energies is smaller (it also does not exceed a factor of 2). At higher energies (480 and 660 Mev) the experimental yields exceed considerably the yields expected from the (p, n) reaction for a gallium target. It can thus be concluded that in our experiments the As^{74} and As^{76} isotopes are produced chiefly in the primary reactions of the type (p, xn). The fact that for As^{76} at higher proton energies the experimental cross sections exceed the expected cross sections considerably can be due to the ever increasing contributions of secondary capture reactions of superbarrier fragments,[1-7] whose number of possible channels is sufficiently large. However, one must not attribute this excess entirely to the contribution of secondary reactions, and one must keep in mind possible peculiarities in the course of the (p, n) reaction on Ge^{76} compared to its course on the Ga^{69} isotope.

The heavier arsenic isotopes (As^{77} and As^{78}) are apparently produced in secondary capture reactions of superbarrier fragments. In the case of the As^{77} isotope both secondary capture reactions of deuterium and tritium, and secondary [p(α, pxn)] reactions are at various proton energies equally possible. Unfortunately direct measurements of the yields of the latter are in general lacking, and it is difficult to separate their contributions exactly. Using the data on the [p(α, xn)] reaction on copper[1] and the ratios of the yields of the (α, pxn) and the (α, xn) reactions on targets close to ours, one can estimate the contributions of the [p(α, pxn)] reactions on some germanium isotopes at 300 MeV. In the case of As^{77} the estimate indicates that the yield of these reactions does not exceed 42—45 percent of the observed yield of the isotope at the corresponding proton energy, and is, if anything, smaller. In view of the fact that the yields of the [p(α, xn)] reactions increase monotonically with increasing proton energy[3,4,7] the contributions of the [p(α, pxn)] reactions for 120-MeV protons should be approximately half as large, and at 660 MeV twice as large as at 300 MeV. Thus, the contribution of competing secondary capture reactions of α particles at 120 MeV is expected to be no more than 20 percent of the observed As^{77} cross section, while at 660 MeV it can completely account for the production of the given isotope. This gives grounds for considering the As^{77} yield at 660 MeV as the upper limit of the sum of the cross sections of the reactions [p(α, p)] and [p(α, p2n)] on the Ge^{74} and Ge^{76} isotopes.

A smaller number of channels takes part in the production of the As^{78} isotope. The latter is chiefly obtained from the secondary capture reactions of superbarrier tritium nuclei. The contribution of the competing capture reaction of α particles of the type [p(α, pn)] is not large. At an energy of 300 MeV it is estimated[1,26] to be ~ 0.003 mb, and according to a much less rigorous estimate[1,20] it amounts to < 0.015 mb, i.e., about 3 percent and not more than 16 percent of the observed As^{78} yield. For proton energies of 120 and 660 MeV the contribution of this competing reaction is estimated to be respectively about 1 and 17 percent of the observed As^{78} yields. Hence it follows that with increasing proton energy a stronger decrease takes place in the yields of the secondary reactions of the capture of deuterium and tritium compared with the observed yields of As^{77} and As^{78}. Direct measurements of the yields of deuterons with kinetic energies > 50 MeV from carbon bombarded with 95,[27] 300,[28] and 660-MeV[10] protons confirm the observed tendency of the As^{77} yields to decrease. However, it is difficult to speak of a strict, quantitative correspondence between data from targets that differ so considerably.

It must be noted that the behavior of yields of secondary capture reactions of deuterium and tritium as a function of the proton energy is the opposite of the behavior of secondary capture reactions of α particles, lithium or beryllium nuclei, which exhibit a characteristic increase of the yield at higher energies.

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

[2] A. Turkevich and N. Sugarman, Phys. Rev. **94**, 728 (1954).

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

[4] Kuznetsova, Mekhedov, and Khalkin, Atomnaya énergiya **4**, 455 (1958).

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

[6] Leffort, Simonoff, and Tarrage, Nucl. Phys. **19**, 173 (1960).

[7] Wang, Kuznetsov, Kuznetsova, and Khalkin, JETP **39**, 230 (1960), Soviet Phys. JETP **12**, 166 (1961).

[8] Wang, Kuznetsov, Kuznetsova, Mekhedov, and Khalkin, JETP **39**, 527 (1960), Soviet Phys. JETP **12**, 370 (1961).

[9] Belyaev, Mal'tseva, Mekhedov, Ming, and Shimchak, JETP **43**, 1129 (1962), Soviet Phys. JETP **16**, 797 (1963).

[10] Azhgireĭ, Vzorov, Zrelov, Meshcheryakov, Neganov, and Shabudin, JETP **33**, 1185 (1957), Soviet Phys. JETP **6**, 911 (1958).

[11] Leffort, Tarrage, Simonoff, and Bobron, J. Phys. Rad. **20**, 959 (1959).

[12] Brink, Katalas, Sharp, Weiss, and Irvine, J. Ann. Chem. Soc. **79**, 1303 (1957).

[13] G. H. Morrison and H. Freiser, Solvent Extraction in Analytical Chemistry, N. Y. 1957.

[14] A. Kyelberg and A. C. Pappas, Inorg. Nucl. Chem. **11**, 173 (1959).

[15] M. Ya. Kuznetsova and V. N. Mekhedov, Izv. Akad. Nauk SSSR, ser. fiz. **21**, 1020 (1957), Columbia Tech. Transl. p. 1021.

[16] Strominger, Hollander, and Seaborg, Rev. Modern Phys. **30**, 585 (1958).

[17] Kuznetsova, Ming, Rybakov, and Khalkin, Radiokhimiya (Radiochemistry) **6**, 755 (1961).

[18] Ball, Fairhall, and Halpern, Bull. Amer. Phys. Soc. **3**, 322 (1958).

[19] I. W. Butler and C. R. Gosset, Phys. Rev. **108**, 1473 (1957).

[20] N. I. Porile, Phys. Rev. **115**, 939 (1959).

[21] R. W. Deutsch, Phys. Rev. **97**, 1110 (1955).

[22] I. W. Meadows, Phys. Rev. **98**, 744 (1955).

[23] I. W. Meadows, Phys. Rev. **91**, 885 (1953).

[24] N. T. Porile, Phys. Rev. **125**, 1379 (1962).

[25] A. A. Caretto Jr. and E. O. Wiig, Phys. Rev. **115**, 1238 (1958).

[26] S. Amiel, Phys. Rev. **116**, 415 (1959).

[27] W. Selove, Phys. Rev. **101**, 231 (1956).

[28] W. H. Hess and B. J. Moyer, Phys. Rev. **101**, 337 (1956).

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284