Letters to the Editor

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Communications should not in general exceed 600 words in length.

Relative Abundances of the Oxygen Isotopes

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ASTON[1] gave the abundance ratios $O^{16}/O^{18}=536$ and $O^{18}/O^{17}=4.2$. Mecke and Childs[2] give for atmospheric oxygen $O^{16}:O^{18}:O^{17}=630\pm20:1:0.2$. Smythe[3] has measured $O^{16}/O^{18}=503\pm10$. A little later Bleakney and Hipple[4] reported values for this ratio clustering around 500 in over a hundred analyzed samples, but gave no estimate of error.

A new determination of the O^{18}/O^{17} ratio has been made, with the 60° mass spectrometer of Nier[5] to separate the singly ionized molecular oxygen ions of masses 32, 33 and 34. Though the present work is to fix more closely the O^{18}/O^{17} ratio which has generally been taken as 5, a check was also made on the value of O^{16}/O^{18}. Commercial oxygen from two different tanks was analyzed several times, ten readings being taken for each analysis. Air samples also were analyzed. As a test for discrimination in the mass spectrometer, runs were made under a wide variety of conditions and at different times. At no time did the O^{16}/O^{18} ratio as computed from the relative heights of the mass 32 and 34 peaks differ by more than 3 percent from 500.

It thus appears that the apparatus does not discriminate appreciably, and that the samples analyzed are representative of the samples analyzed by Smythe and Bleakney and Hipple. In each of ten separate measurements of the O^{18}/O^{17} ratio, a complete mass spectrum of the 33 peak and its adjacent valleys has been taken in order to obtain an accurate determination of the background. From the ratio of the deflections corrected for background, the average value of O^{18}/O^{17} is 4.9 ± 0.2.

From the values $O^{16}/O^{18}=500\pm15$ and $O^{18}/O^{17}=4.9\pm0.2$, one is able to calculate the physical-to-chemical mass scale conversion factor to be 1.000275 ± 0.000009. By far the larger part of this uncertainty is due to the possible error in the ratio O^{16}/O^{18}.

My thanks are due to Dr. A. O. Nier for his suggestion of the problem and for aid in working it out.

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Radioactive Isotopes of Germanium

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WE have studied the radioactive isotopes of germanium produced by deuteron and neutron bombardment of germanium, fast neutron bombardment of arsenic and selenium, and deuteron bombardment of gallium. We were able to make isotopic assignments for four germanium periods and to establish the existence of a pair of isomers in Ge^{71}.

Ge^{71}: 40±2 hours and 11 days.—Sagane[1] has reported a 26-hour and a 6-day period from both slow and fast neutron bombardment of germanium; without chemical separation he assigned the positron active 26-hour activity to Ge^{71} and suggested that the 6-day period may be due to a gallium isotope. In an alpha-particle bombardment of zinc, Mann[2] obtained chemically identified germanium isotopes with half-lives of 37 hours and 195 days and an intermediate period whose half-life, although not given by Mann, turns out, from his curves, to be 11 days. These three periods have to be distributed between Ge^{67}, Ge^{69}, and Ge^{71}.

Bombarding gallium with 8-Mev deuterons we found in the germanium fraction a 40±2-hour positron-emitter and an 11-day activity. These are almost certainly due to Ge^{69} or Ge^{71}, produced by the d, $2n$ reaction from the only stable gallium isotopes, Ga^{69} and Ga^{71} (d, n reactions would lead to stable germanium isotopes). But since the bombardment of germanium with 16-Mev deuterons also produced a 40±2-hour and an 11-day germanium period, both periods must be assigned to Ge^{71}, formed by the d, p reaction from Ge^{70}. Absorption measurements show that the positrons of the 40-hour activity have an energy of 1.2 Mev, which agrees with Sagane's 1.15 Mev for his 26-hour period.[3] The 11-day activity emits particles of about 0.6-Mev energy, and no gamma-rays. The absence of annihilation radiation indicates that very few, if any, positrons are emitted; the particles may be conversion electrons following K-electron capture.

Ge^{75}: 89±2 minutes.—In slow neutron bombardments of germanium Amaldi *et al.*[4] obtained a 30-minute period, while Sugden[5] reported a 2-hour activity. Pool, Cork, and Thornton[6] found a 1.3-hour activity in fast neutron bombardment of germanium. Sagane[1] found a negative beta-emitter of 81 minutes half-life in slow and fast neutron and in deuteron bombardments of germanium and assigned it to Ge^{75}. No chemical analyses were reported in any of the experiments quoted. We found that 16-Mev deuteron bombardment of germanium produced a 90-minute period in the germanium fraction. To fix the assignment we bombarded arsenic with fast neutrons. The germanium fraction decayed with a period of 89±2 minutes over about 10 half-lives. This activity must be formed from the only stable arsenic isotope, As^{75}, by the n, p reaction, and must therefore be due to Ge^{75}. We also found the same period (about 90 minutes) in the germanium fractions of fast neutron bombardments of germanium and selenium, where Ge^{75} can be formed by the reactions $Ge^{76}(n, 2n)Ge^{75}$

and $Se^{78}(n, \alpha)Ge^{75}$, respectively. Absorption measurements show the maximum energy of the beta-particles from the 89-minute activity to be 1.2 ± 0.1 Mev, in agreement with Sagane's value of 1.1 Mev for his 81-minute period.[3] Gamma-rays are present but their intensity relative to the beta-particles is rather low.

Ge^{77}: 12 hours.—Sagane[1] reported a negative beta-emitter of 8 hours' half-life from slow neutron and deuteron bombardments of germanium and assigned it to Ge^{77}, because fast neutrons had produced very little of this activity. Our experiments confirm this assignment but show the half-life to be about 12 hours. This period, chemically identified as due to germanium, was produced by the *d, p* reaction in the bombardment of germanium with 16-Mev deuterons and by the *n*, α reaction in the fast neutron bombardment of selenium. These results, combined with the fact that the 12-hour period did not appear in fast neutron bombardments of either germanium or arsenic, make its assignment to Ge^{77} certain. The growth of As^{77} (90 days)[7] from this activity has not yet been investigated.

Ge^{69}.—In addition to the periods discussed, Sagane[1] reported a positron-emitting 29-minute activity which he obtained from fast neutrons on germanium only and therefore assigned to Ge^{69}. We failed to observe this period from the reactions $Ge^{70}(n, 2n)Ge^{69}$ and $Ga^{69}(d, 2n)Ge^{69}$ and Mann[2] did not find it from the reaction $Zn^{66}(\alpha, n)Ge^{69}$. These facts lead us to reject Sagane's assignment. The 195-day period reported by Mann[2] may be due to Ge^{69}.

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Radioactive Isotope of Protoactinium

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HAHN and Strassmann[1] have recently pointed out that the radioactivity of 25 days half-life assigned[2] to $_{91}Pa^{233}$, originally reported by Meitner, Strassmann and Hahn[2] to be the daughter of the 26-minute $_{90}Th^{233}$ produced by neutron capture in thorium, might actually be due to an isotope of zirconium. There is a 26-day zirconium[1] formed in the fission of uranium by neutrons, and hence probably also in the fission of thorium by neutrons. Since Meitner, Strassmann and Hahn[2] used zirconium as the carrier material for their radioactive substance, the question arises as to whether their 25-day radioactivity from thorium plus neutrons might be due to an isotope of zirconium rather than protoactinium.

We have investigated this point and our experiments definitely show that, in agreement with the original conclusions of Meitner, Strassmann and Hahn,[2] slow neutron bombardment of thorium produces a protoactinium of approximately 25 days half-life.[3] About 10 g of thorium nitrate, encased in a large paraffin block, was exposed for an hour to the neutrons produced during the bombardment of phosphorus with 100 microamperes of deuterons in the 60-inch cyclotron. The thorium was dissolved and precipitated from acid solution as the iodate, after the addition of appropriate carriers for the fission products, and then, after redissolving the thorium iodate in concentrated hydrochloric acid, successive precipitations of zirconium phosphate were performed. This showed that a 25-day period was growing from a parent of approximately 26 minutes half-life, presumably $_{90}Th^{233}$. The cross section for the production of the 25-day activity was very much greater than that of any of the fission products (e.g., antimony, tellurium, iodine, etc.). The absence in the zirconium phosphate fractions of the 17-hour zirconium, known[4] to be produced in the fission of uranium by neutrons, suggests that the amount of radioactive zirconium formed from fission in this experiment was negligible.

Final proof that the 25-day activity is an isotope of protoactinium, and not of zirconium, was obtained by separating protoactinium and zirconium by fractional crystallization[5] of zirconium oxychloride from hydrochloric acid solution. A sample of greater intensity was prepared, zirconium carrier was added and the sample was purified carefully and converted to zirconium oxychloride. The fractional crystallizations showed that the specific activities (activity per milligram ZrO_2) of the first fractions were of the order of 5 percent of the specific activity of the over-all sample, showing an accumulation of the activity in the mother liquor as should be the case for protoactinium. A sample of 17-hour zirconium was prepared (from uranium plus neutrons) and subjected to a similar series of fractional crystallizations; in this case the specific activities of all the fractions were the same as that of the over-all sample, as was to be expected.

Absorption measurements show the presence of a gamma-ray, and an upper limit for the beta-rays of about 0.4 Mev.

A search for $_{92}U^{233}$ activity from the decay of $_{91}Pa^{233}$ was made by chemically separating uranium. This uranium fraction was inactive with respect to beta-particle emission. The question of alpha-radioactivity is being investigated. $_{90}Th^{233}$ and $_{91}Pa^{233}$ are members of the $4n+1$ series of radioactive elements (the series which is missing among the natural radioactivities).

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