Radioactivities of germanium, arsenic, gallium and zinc

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Abstract
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Disciplines
Chemistry
RADIOACTIVITIES OF GERMANIUM, ARSENIC, GALLIUM AND ZINC

By
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June 22, 1950

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Abstract

Radio activities were produced by the fast (pile) neutron bombardment of germanium. Chemical separations followed by decay and absorption measurements yielded the following results. Eleven-day Ge$^{71}$ decays with the emission of positrons with maximum energy of 0.65-Mev Ge$^{77}$ with 12 hour half-life emits 0.31-Mev gamma-rays. The half-life of As$^{77}$ is 38.0±0.5 hours and it emits 0.68 Mev $\beta$-particles and two $\gamma$-rays. The following new gallium and zinc activities were observed, but not assigned definite mass numbers. A 14.5 day gallium activity emits $\beta$-particles with energies of 0.29 and 0.71 Mev, 34 Kev $\gamma$-rays and possibly X-rays; a 53 hour gallium activity gives off 1.9 Mev $\beta$ particles and 1.63 Mev $\gamma$-rays; a 14-day zinc activity decays with the emission of 2.1 Mev $\beta$ particles and 40 Kev $\gamma$ rays.
Radioactivities of Germanium, Arsenic, Gallium, and Zinc*

by Warren Heiman and Adolf Voigt

Introduction

The fast neutrons produced in fission have sufficient energy to induce some reactions other than the \( n, \gamma \) process most frequently observed with the moderated neutrons used in pile bombardments. In order of increasing energy requirements and consequent lower probability the \( n, p, n, \alpha \), and \( n, 2n \) reactions might be observed. The desired radioactivities can be found more readily if the thermal component of the neutron spectrum is filtered out. Consequently cadmium wrapped samples of materials are frequently irradiated in order to study these activities.

Since the atomic number of germanium is low the positive particles, the proton and the alpha particles, would not require too high an energy to be ejected, hence, germanium was chosen as an element for study. Also it was felt possible that certain discrepancies in the reported activities for germanium could be corrected and certain new activities might be produced. By means of the fast neutron bombardment of germanium, rather high specific activities may be obtained for the radioactive isotopes of gallium, zinc, and arsenic, because the germanium may be completely separated from them by use of a special distillation apparatus.

The unstable nuclides possibly produced by these reactions would be Zn 69, 71, and 73 by \( n, \alpha \); Ga 70, 72, 73, 74, and 76 by \( n, p \); Ge 69, 71, and 75 by \( n, 2n \) in addition to Ge 71, 75, and 77 by \( n, \gamma \). Daughter products could also be produced.

*Work performed under Contract No. W-7405 eng 82.
**Chemical Procedure**

Spectroscopically pure, one gram samples of GeO$_2$ were bombarded with neutrons in the nuclear reactor of the Argonne National Laboratory. Each sample was contained in a silica tube which was wrapped with approximately 1/32 inch cadmium foil to absorb the thermal neutrons. The lengths of the bombardments varied from five days to two months.

Upon receipt of a bombarded sample, the radioactive GeO$_2$ was dissolved in dilute NH$_4$OH. This solution was then made strongly acid with concentrated HCl. Zinc, arsenic, and gallium carriers were then added and the solution was transferred to a special apparatus for the distillation of GeCl$_4$ under an atmosphere of Cl$_2$. The GeCl$_4$ was completely removed by distillation and precipitated as GeS$_2$.

The solution remaining in the distilling flask contained arsenic, gallium, and zinc. The arsenic was precipitated from 9 N HCl as the sulfide. The arsenic was filtered out and the filtrate contained gallium and zinc. The filtrate was made 7 N in HCl and the gallium was extracted with isopropyl ether. The gallium was precipitated as Ga(OH)$_3$ and the zinc which remained in the aqueous phase was taken down as ZnNH$_4$PO$_4$. The procedure for gallium and zinc was modified in one of the bombardments in order to detect any possible tin impurity. Tin was added with the other carriers and subsequently separated by ether extraction. It was precipitated as Sn(OH)$_4$ after the separation.

**Germanium**

Activities which followed germanium chemistry immediately after
the samples arrived at Ames had half-lives of 31 hours and 11 days plus a low intensity 8 hour component. (Fig. 1). Reseparation 5 days after irradiation produced an arsenic fraction with a half-life of 40 hours while the sole half-life in germanium was 11 days. These results indicate that at the time of the first separation the 12 hour Ge$^{77}$ was present, growing into its 40 hour As$^{77}$ daughter after the separation. The resulting half-lives were observed as ~ 8 hours and 31 hours since the decay curve was not resolvable into its 12 hour and 40 hour components. By the time of the second separation the 12-hour activity had decayed out leaving only the 11-day Ge$^{71}$ in the germanium fraction and the 40 hour activity in the arsenic.

A 40-hour activity is also reported$^2$ for Ge$^{69}$. This would have to be produced by an $n_22n$ reaction in these experiments which is not highly probable at these neutron energies. In the present experiments its presence would have been masked by the 40-hour As$^{77}$. All that can be said is that it was not produced in sufficient yield to be observed when the second separation was run 5 days after the irradiation. However, if, as has previously been reported$^3$, the 40-hour and 11-day germanium activities were isomers of Ge$^{71}$, the former would be expected in large enough amounts to be detectable 5 days after irradiation. Consequently, our evidence confirms the present assignment. Supporting evidence was obtained by considering the sign of the particles as determined by counting in a magnetic field. The 40-hour activity showed a large ratio of negative to positive particles in agreement with its
Fig. 1 — Decay curve of Germanium.

Fig. 2 — Aluminum absorption curve of germanium three days after bombardment.
assignment to As\textsuperscript{77} rather than Ge\textsuperscript{69} or Ge\textsuperscript{71} which would be positron emitters. The 11-day germanium, on the other hand, gave a higher positive than negative count showing that it decays to gallium.

By absorption measurements it was shown that a $\beta$-ray with a range of 1.0 g/cm\textsuperscript{2} of aluminum, (Fig. 2) energy 2.1 Mev, decayed out rapidly and hence was most likely associated with the 12 hour Ge\textsuperscript{77}. This agrees with the previous value of 2.0 Mev\textsuperscript{4} for the Ge\textsuperscript{77} $\beta$-ray. Also associated with this decay was a gamma ray with a half-thickness in aluminum of 6.4 g/cm\textsuperscript{2}, corresponding to an energy of 0.3 Mev. The 11-day germanium $\beta$-ray had a range of 230 mg/cm\textsuperscript{2}, energy 0.65 Mev. (Fig. 3).

**ARSENIC**

The only activity observed in the arsenic fractions was As\textsuperscript{77} for which the half-life was measured as 38 $\pm$ 0.5 hours, (Fig. 4) compared to the previously reported value of 40 hours\textsuperscript{4} Feather analysis of an aluminum absorption curve (Fig. 5) gave a range of 246 mg/cm\textsuperscript{2} (Fig. 6), corresponding to an energy of 0.68 Mev in agreement with the value of 0.7 listed in the Trilinear Chart\textsuperscript{5}.

A portion of the arsenic sample was mounted for the thin-lens magnetic $\beta$-ray spectrometer and the $\beta$-ray spectrum was measured by E. N. Jensen of this laboratory\textsuperscript{6}. The value so obtained was 0.679 Mev, showing that the Feather method was reliable. The $\beta$-ray spectrum showed a deviation from the straight-line Fermi plot at low energies, indicating the possible presence of a component with an energy of 0.36 Mev and a yield 12% as great as that of the major component. However, it is Dr.
Fig. 3 — Decay curve of germanium obtained from the second separation.

Fig. 4 — Decay curve of arsenic obtained from the second separation.
Fig. 5 — Aluminum absorption curve of arsenic.

Fig. 6 — Feather Analysis of Arsenic.
Jensen's current opinion that this is an artifact due to the relatively thick sample (0.8 mg/cm²) used in this case, and that a thin source would probably not show this.

Conversion lines from gamma rays were not seen in the results from the beta spectrometer, but lead absorption curves (Fig. 7) showed two \( \gamma \)-rays of fairly low intensity. Half-thickness values in lead were 85 mg/cm² and 3.4 g/cm² corresponding to energies of 0.05 and 0.42 ± 0.04 Mev.

GALLIUM

Three distinct gallium radioactivities were produced by the fast neutron bombardment of germanium. Decay periods of 15 hours, 53 hours and 14.1 days are shown in Figure 8. A decay curve, followed over a period of four half-lives, indicates that the long-lived activity has a half-life of 14.5 days (Figure 9). The 15-hour activity was identified with 14-hour Ga⁷²; the other two do not correspond to known gallium isotopes.

Several absorption curves in aluminum (Fig. 10) and copper (Fig. 11) were taken at intervals during the course of the decay. The aluminum curves were subjected to Feather analysis. It was observed that 0.29 and 0.71 Mev \( \beta \)-particles and a 34 Kev \( \gamma \) ray are associated with the 14.5 day activity. The portion of the curves that is interpreted as the 0.71 Mev \( \beta \) might instead be due to a germanium X-ray, though its intensity relative to the other components seems too high for this. The 53-hour activity possesses a 1.9 \( \beta \) ray and a 1.63 Mev \( \gamma \) ray. The
Fig. 7—Lead absorption curve of arsenic.

Fig. 8—Decay curve of gallium.
Fig. 9 — Decay curve of gallium with some 38 hour arsenic present.

Fig. 10 — Aluminum absorption curve of gallium.
Fig. 11—Copper absorption curve of gallium.

Fig. 12—Decay curve of zinc.
β-particles were found to be negatively charged by observation of the counts taken in a magnetic field.

Consideration of the β-ray energies involved indicates that the 14.5-day activity was either a nuclear isomer of Ga\textsuperscript{70} or unreported Ga\textsuperscript{74}. The 53-hour activity may be a nuclear isomer of Ga\textsuperscript{70} or Ga\textsuperscript{73}, or it may be caused by Ga\textsuperscript{74}. The assignment of any of the activities to Ga\textsuperscript{74} appears to be somewhat questionable on the basis of following observations.

Sagane, Miyamoto and Ikawa\textsuperscript{7} observed a 9-day gallium activity as a result of bombarding germanium with deuterons and assigned it to Ga\textsuperscript{74}. Siegel and Glendenin\textsuperscript{8} did not observe this 9-day gallium activity as a fission product of uranium. McCown, Woodward, and Pool\textsuperscript{2} attempted to produce Ga\textsuperscript{74} by the \(d\alpha\) reaction on germanium metal enriched to 70 per cent Ge\textsuperscript{76}. However, the only gallium activity observed was the 14-hour period of Ga\textsuperscript{72}. It can be seen by considering the isotopic abundances, the expected stability of the nuclei using the Bohr-Wheeler type of calculation and the fission yield distribution that half-lives of this length are less probable for Ga\textsuperscript{76} than for Ga\textsuperscript{74}. In the light of this conflicting evidence we are reluctant to make any definite assignments until further work is carried out.

The possibility of the 53-hour or 14.5-day activities being due to the presence of radioactive impurities was shown to be highly unlikely through consideration of the amounts of the impurities known to be present.

**Zinc**

The zinc fraction obtained from the fast neutron bombardment of
germanium yielded half-lives of 14 hours and 14 days (Figure 12). Beta-particles were detected with energies of 0.91 Mev and 2.1 Mev (Figure 13). There were two $\gamma$-rays of ~0.2 Mev and 0.04 Mev (Figure 14). The $\gamma$-ray energies are somewhat uncertain because of the low intensity of the activity, however the beta-gamma ratio was 110, indicating that these were $\gamma$-rays and not bremsstrahlung.

The 0.91 Mev $\beta$ and the ~0.2 Mev $\gamma$ were shown to decay with a 14 hour half-life, corresponding to the isomeric transition (0.44 Mev $\gamma$) and $\beta$ ray (10 Mev) reported for Zn$^{69}$. The characteristics and assignment of the 14 day activity remain. Chemical evidence is strong against its being due to an impurity, though this remains a slight possibility. The 2.1 Mev $\beta$ and 40 kev $\gamma$ ray belong to the 14 day activity. The $\beta$ rays were shown to be negatively charged by counting in a magnetic field.

An $n, \alpha$ reaction in germanium can produce Zn$^{69}$, Zn$^{71}$ and Zn$^{73}$. Of these Zn$^{69}$ is the 14 hour activity discussed above and Zn$^{71}$ is reported to emit a $\beta$-ray of 2.1 Mev energy with a half-life of 2.2 minutes. From this information it is seen that the most probable assignment of the 14-day activity would be the unreported Zn$^{73}$ or a nuclear isomer of 2.2 minute Zn$^{71}$. Of these two, general energy considerations could make it appear unlikely that such a long half-life could be assigned to Zn$^{73}$ since the heaviest stable isotope is Zn$^{70}$. Also fission product studies indicate that the half-life of Zn$^{73}$ must be less than 2 minutes. Assignment of the 14 day activity to a nuclear isomer of Zn$^{71}$ might be considered to have some basis since 2.1 Mev is also reported as the
Fig. 13 — Aluminum absorption curve of zinc.

Fig. 14 — Copper absorption curve of zinc.
energy of the 2.2 minute Zn$^{71}$. The low abundance of Zn$^{70}$ (0.6%) possibly combined with a low cross section for neutron capture by this isotope would make a 14 day activity hardly observable in neutron irradiated zinc compared to the fairly high activity of 250 day Zn$^{65}$ obtained from Zn$^{64}$ (abundance 49%, cross section $0.5 \times 10^{-24}$ cm$^2$). Experiments are in progress to check the possibility of this isomerism.

**SUMMARY**

The following table summarizes the results obtained in the present work.

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Half-life</th>
<th>Type of Radiation</th>
<th>Energy of radiation in MeV</th>
<th>Particles</th>
<th>γ-rays</th>
</tr>
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<tbody>
<tr>
<td>Ge$^{71}$</td>
<td>11 days</td>
<td>$\beta^+\alpha$</td>
<td>0.65</td>
<td>None</td>
<td></td>
</tr>
<tr>
<td>Ge$^{77}$</td>
<td>12 hours</td>
<td>$\beta^-\gamma$</td>
<td>2.1</td>
<td>0.31</td>
<td></td>
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<tr>
<td>As$^{77}$</td>
<td>38 hours</td>
<td>$\beta^-\gamma$</td>
<td>0.68</td>
<td>0.42, 0.05</td>
<td></td>
</tr>
<tr>
<td>Ga$^{70}$ or Ga$^{74}$</td>
<td>14.5 days</td>
<td>$\beta^-\gamma$</td>
<td>0.29, 0.71</td>
<td>0.034</td>
<td></td>
</tr>
<tr>
<td>(Zn$^{71}$)</td>
<td>14 days</td>
<td>$\beta^-\gamma$</td>
<td>2.1</td>
<td>0.040</td>
<td></td>
</tr>
</tbody>
</table>


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