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ALPHA DECAY PROPERTIES OF NEUTRON DEFICIENT ISOTOPES OF EMANATION

Kalevi Valli, Matti J. Nurmin, and Earl K. Hyde

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ABSTRACT

Isotopes of emanation (element 86) with 126 or fewer neutrons were prepared at the Heavy Ion Linear Accelerator by bombardment of platinum, gold, mercury, and thallium targets with $^{16}$O, $^{14}$N, and $^{12}$C nuclei. Several techniques were developed for isolation of the gaseous products. Si(Au) surface-barrier detectors were used to measure alpha particles emitted by emanation isotopes and by their polonium and astatine daughters. Mass number assignments of new isotopes of mass 201 through 205 were based on genetic relationships and on excitation functions. The probable nature of the isomerism observed in $^{203}$Em is discussed. Properties of isotopes with mass 206 through 212 were confirmed.
I. INTRODUCTION

There exist published data on isotopes of the element emanation with 126 or fewer neutrons with half lives ranging from 16 hours (\(^{211}\text{Em}\)) down to three minutes (\(^{204}\text{Em}\)) (Table I). These isotopes can be prepared by spallation reactions in thorium bombarded with high energy protons or by bombardment of elements below polonium with heavy nuclei such as nitrogen. The available knowledge on these isotopes is summarized by Hyde, Perlman, and Seaborg.\(^6\)

In the work reported here the study of emanation isotopes has been resumed with nuclear reactions induced by heavy nuclei accelerated in the Berkeley Heavy Ion Linear Accelerator (HILAC). New bombardment and sample preparation techniques and improved semiconductors have made it easier to investigate short lived alpha-emitting isotopes. This new work confirms the previously reported information on isotopes from mass 206 to mass 212 and provides information on several new isotopes from mass 205 to mass 201.

These new data are useful in connection with the determination of nuclear masses of very light nuclei and in the systematics of decay energies in this region of the nuclidic chart. The discovery of isomeric forms of \(^{203}\text{Em}\) is interesting in connection with an "island of isomerism" systematically occurring in the light odd-A isotopes of platinum, mercury, lead, polonium, and emanation.
II. EXPERIMENTAL TECHNIQUES

The nuclear reactions written below were used to produce the emanation isotopes. All involve the formation of a compound nucleus which begins its de-excitation by the emission of three or more neutrons. At the full bombarding energy of 10.3 MeV per nucleon the compound nucleus has enough excitation energy to evaporate ten or more neutrons.

\[
\begin{align*}
203,205_{\text{Tl}} + ^{14}_{\text{N}} & \rightarrow \left[ ^{217,219}_{\text{Ra}} \right] \text{-xn followed by } \alpha \text{ decay} \quad \text{Em isotopes below 213} \\
203,205_{\text{Tl}} + ^{12}_{\text{C}} & \rightarrow \left[ ^{215,217}_{\text{Fr}} \right] \text{-xn followed by } \beta^c \text{ decay} \quad \text{Em isotopes below 214} \\
80_{\text{Hg}}(\text{nat.mix.}) + ^{12}_{\text{C}} & \rightarrow \left[ ^{215, \text{and less }}_{\text{Em}} \right] \text{-xn} \quad \text{Em isotopes below 214} \\
197_{\text{Au}} + ^{16}_{\text{O}} & \rightarrow \left[ ^{213}_{\text{Fr}} \right] \text{-xn followed by } \beta^c \text{ decay} \quad \text{Em isotopes below 211} \\
197_{\text{Au}} + ^{14}_{\text{N}} & \rightarrow \left[ ^{211}_{\text{Em}} \right] \text{-xn} \quad \text{Em isotopes below 209} \\
^{78}_{\text{Pt}}(\text{nat.mix.}) + ^{16}_{\text{O}} & \rightarrow \left[ ^{214, \text{and less }}_{\text{Em}} \right] \quad \text{Em isotopes below 212}
\end{align*}
\]

For those emanation isotopes with half lives longer than a minute one successful technique was to bombard metallic targets of platinum, gold, or
thallium, with an appropriate heavy ion beam for a few minutes and then insert the bombarded metal in the apparatus drawn in Fig. 1. The apparatus was evacuated and the target metal was quickly melted by torch or by an induction coil connected to a high frequency power source. The glow discharge technique described by Momyer and Hyde was used to deposit a fraction of the emanation radioactivity onto a small piece of aluminum foil. Our electrodes were tungsten wires about 1 cm apart inside the glass tube. A piece of 0.005-inch aluminum foil was clamped to the cathode. A variable high voltage power supply was connected to the electrodes through a 0.1 megohm resistor and the current was adjusted to a few milliamperes to induce a purple glow discharge. If the pressure in the tube was too low to support the discharge, air was introduced through a controlled leak. A discharge time of 20 to 60 seconds was sufficient for collection of a sample with a decay rate of several million disintegrations per minute. The efficiency of the deposition process is not known but apparently is in the range of 1 to 10 percent. The main advantage of this technique was that it results in excellent thin samples for alpha particle and alpha gamma coincidence measurements.

A second method of sample preparation is shown in Fig. 2. The bombarded target metal was dropped into the quartz tube on the left side of the apparatus, the system was evacuated, and the target material was melted. The emanation thus set free was allowed to expand into the counting chamber where the alpha particles emitted by the gas were observed with a semiconductor detector. Separation times of less than 40 seconds from the end of irradiation to the beginning of alpha spectroscopy were consistently achieved with this method.
The on-line apparatus shown in Fig. 3 was used for the study of emanation isotopes with half lives of one minute or less. A strip of an 0.001-inch thick gold or platinum foil was mounted in the evacuated target chamber. The ion beam of 0.01 to 1.5 microamperes entered the chamber through an 0.00015-inch nickel window. At low beam currents the emanation isotopes produced were released by heating the target strip with electric current during or after the bombardment. At high beam currents the heat developed by the beam itself was sufficient to release the emanation. The target chamber was connected by a short tube to a counting chamber identical to the one diagrammed in the right side of Fig. 2. When bombardment times were longer than a few minutes the valve between the counting chamber and the pump was kept slightly open in order to counteract a slow build-up of pressure. The energy of the α particles emitted by the gaseous activity in the counting chamber was determined by operation of the semiconductor detector and multichannel pulse height analyzer in the period between beam pulses. The beam pulse had a width of 5 milliseconds and a repetition rate of 40 per second.

The apparatus in Fig. 3 was also used for measuring excitation functions of the new activities. In a series of experiments the beam energy was varied by inserting various thicknesses of aluminum absorber in front of the target chamber as indicated in Fig. 3. Northcliffe's range energy curves were used to compute the 14N energy as a function of aluminum foil thickness. The target consisted of thin gold leaf (2.5 mg/cm²). Gold was the best choice for the excitation function work because it has only one stable isotopes. Data were collected with the semiconductor detector operating continuously between
beam pulses until a predetermined amount of beam had passed through the target. For this purpose the counter chamber was suitably insulated for use as a Faraday cup.

Alpha energies were measured with Si(Au) surface barrier detectors manufactured by Nuclear Diodes Inc. and Ortec. In most experiments detectors with 25 mm$^2$ area and resolution of 15 to 20 keV were used. A preamplifier, an amplifier, and a biased amplifier, all of Lawrence Radiation Laboratory design, were used. Pulse height analysis of the output pulses was done with Tullamore 800 channel analyzer. Energy scales were calibrated with alpha standards made from the active deposit of the thorium and actinium natural decay series. Specifically, the following values, taken from the work of Rytz, were used as energy standards: $^{211}\text{Bi}$, 6.6222 and 6.2775 MeV; $^{212}\text{Bi}$, 6.0506 and 6.0898 MeV; and $^{212}\text{Po}$, 8.785 MeV. Polonium-210, 5.3048 MeV, was also used. Calibration sources were placed in the same chamber with the sample to be measured so that there could be no question concerning gain shifts between time of measurement of sample and standard.

Half lives of short-lived alpha activities were measured by use of special timing circuits similar to those described by Macfarlane and Griffioen. A HILAC ion source control unit was used to produce beam bursts on the target during a period of a few seconds. The same control unit tripped a mercury relay which regulated the flow of an electrical heating current through the metallic target during the bombardment period. This insured rapid release of the emanation activity. After that the beam was automatically turned off, and a timing control circuit activated the Tullamore 800 channel analyzer to record 100-channel spectra of the decaying $\alpha$-activities in eight successive time periods.
The time devoted to each 100-channel spectrum could be chosen to meet the demands of the experiment, and a new beam burst was automatically started at the end of the spectroscopy period. A sufficient number of sequences were run to insure statistical significance of the results.

In the early stages of the work we investigated the use of stearate salts as target materials. Salts of stearic acid have a porous crystal structure and hence a high tendency to allow the diffusion of a gaseous atom out of the crystal interior. In the early decades in the study of natural radioactivity this "emanating power" was carefully studied with radon, actinon, and thoron. We prepared stearate salts of erbium, platinum, mercury, thallium, and lead by precipitating them from warm aqueous solutions of soluble sodium stearate with a slight excess of the appropriate heavy metal solution. The precipitates were carefully washed, dried, and out-gassed in vacuum. For use as a target the material was used as a loose powder or was pressed with a steel plunger into thin solid discs or bricks against the back wall of the chamber shown in Fig. 3. The emanation activity formed as a result of nuclear reactions in the interior of the crystals diffused out of the target and into the counting chamber. This method was successful in a number of experiments, but it proved to have no advantage for our purpose over the use of thin metal foils and it had a few drawbacks. The beam current had to be limited to less than 0.05 microampere to prevent excessive damage to the compounds resulting in gas evolution which raised the pressure in the system, impairing the energy resolution. Nonetheless, the use of stearates or similar compounds may prove of use in future experiments with target elements which are not available in metallic form.
III. RESULTS

A. Confirmation of Previous Results on Emanation-208 through Emanation-212

Figure 4 shows the α activity present in an emanation sample isolated from a thick thallium target bombarded for 45 minutes with a 300 nanocurie current of 140 MeV 14N nuclei. The emanation sample was deposited on aluminum by the glow discharge technique. Figure 4 represents 20 minutes of data collection starting about 60 minutes after the bombardment. Other spectra were taken at earlier and later times after bombardment. Energy values and half lives were obtained for the individual peaks in order to confirm the presence of 212$^{\text{Em}}$, 211$^{\text{Em}}$, 210$^{\text{Em}}$, 209$^{\text{Em}}$, and 208$^{\text{Em}}$ as given by the labels in the figure. Our energy determinations with a semiconductor detector are not as precise as those of Momyer, Asaro, and Hyde made in a magnetic spectrograph and we have used their values in Fig. 4. Our results verified that this group of emanation isotopes could be prepared in good yield in reactions induced by complex nuclear projectiles and confirmed the reliability of our sample preparation and α-measurement techniques before we turned to a study of isotopes of lighter mass and shorter half life.

B. Details on Emanation-211 and Emanation-210

We prepared samples of 16-hour 211$^{\text{Em}}$ by bombardment of millimeter thick discs of thallium metal with 126-MeV 12C nuclei for a 2-hour period. Several hours after the bombardment the thallium was melted in the apparatus sketched in Fig. 1 and a portion of the emanation activity was deposited on an aluminum foil by the glow discharge technique. The α spectrum of a freshly prepared sample about 4 hours after the bombardment was dominated by the 6.037-MeV α particle of 2.7 hour 210$^{\text{Em}}$. This half life was confirmed by repeated measurements.
of the \( \alpha \) spectrum over a 24-hour period. Alpha particles of 16-hour \( ^{211} \text{Em} \) become prominent about one day after the bombardment, as shown in Fig. 5. The previously reported groups at 5.847, 5.779, and 5.613 MeV were seen. The 5.847-MeV peak was not resolved from the 5.862-MeV \( ^{211} \text{At} \) daughter peak which, at the time this spectrum was obtained, had grown to equilibrium in the sample.

Our singles spectra add no new information to that obtained by Momyer, Asaro, and Hyde\textsuperscript{3} in a magnetic spectrometer.

Figure 5 also shows an \( \alpha \gamma \) coincidence spectrum obtained in a 7-hour run starting 15 hours after the bombardment. In this experiment \( \gamma \) rays passing through the 5-mil aluminum backing of the sample were detected in a 3 \( \times \) 3 inch sodium iodide crystal coupled to a photomultiplier tube. Alpha pulses from the semiconductor detector were analyzed only if they were coincident with \( \gamma \) ray pulses of any energy above about 20 keV. As expected, the 5.847 MeV ground state transition was absent from the coincidence spectrum while the 5.779 and 5.613 MeV peaks were prominent. The reversal in the intensity ratio of these two peaks in the coincidence spectrum compared to the singles spectrum is a result of the high L- and M- conversion coefficients for the 68.8 keV E2 transition which follows the 5.779 MeV \( \alpha \) particle.

The conversion process also is the cause of the bump at 5.672 MeV in the coincidence spectrum. The L-electrons of the 68.8 keV transition have energies of 52 to 55 keV. These electrons can provide additional ionization in the detector to add to the ionization from the 5.613 MeV \( \alpha \)-particles, resulting in a total effect equivalent to an \( \alpha \)-particle of about 5.67 MeV. The \( \gamma \) gate signal to trip the coincidence circuit is provided in this case by the 169 keV transition. See the decay scheme in Fig. 5.
The peaks appearing at 6.895 and 6.568 MeV are low intensity $\alpha$ groups of $^{211}$Po, the electron capture daughter of $^{211}$At. Fine structure peaks were looked for in the decay of $^{210}$Em by $\alpha \gamma$ coincidence measurements made at a time when $^{210}$Em was a prominent part of the singles spectrum, but no evidence was found for any complex structure.

C. General Remarks on Light Isotopes of Emanation

Some typical spectra taken on emanation samples from Hg+C, Au+N, and Pt+O bombardments are shown in Figs. 6, 7, and 8, respectively. In these experiments full energy projectiles impinged on thick targets and the short lived emanation isotopes were collected continuously and counted between beam pulses.

The most definitive information came from excitation function experiments done with nitrogen ions and thin gold targets. A series of 24 experiments was performed in which the $^{14}$N beam was reduced in energy by a stack of 0.00025 inch aluminum foils. Figure 9 shows the spectra obtained at four beam energies. Striking changes occur in the spectra as the beam energy is decreased. These changes are most apparent when the number of $\alpha$-particles in each distinct peak is plotted as a function of beam energy. Figure 10 shows such excitation curves for the emanation isotopes.

Some of the observed $\alpha$-energies and half lives corresponded to astatine or polonium daughter activities of known mass number. The presence of these isotopes helped in the assignment of mass numbers to the unknown emanation activities. A comparison of the yield versus beam energy curves of the parent and daughter activities was particularly helpful in this respect. For this purpose it was necessary to be explicit about the properties assumed for the light
astatine and polonium nuclides because there has been confusion in the published literature about them. Table II summarizes our choice of properties for the astatine and polonium isotopes.

D. Emanation-207

An alpha energy of 6.12 MeV was assigned to $^{207}\text{Em}$ by Stoner and Hyde. We prepared this isotope relatively free of 6.141 MeV $^{208}\text{Em}$ by bombarding thin targets of gold with 75 MeV $^{14}\text{N}$ ions. From these experiments the alpha energy of $^{207}\text{Em}$ was determined to be 6.135±0.003 MeV. To confirm the assignment of this peak to $^{207}\text{Em}$, we compared the excitation function of this activity to that of 5.752 MeV $^{207}\text{At}$ and found them to be similar.

E. Emanation-206 and Emanation-205

Stoner and Hyde assigned a 6.25-MeV $\alpha$ particle with a half life of 6.2±0.5 minutes to $^{206}\text{Em}$. We produced this activity in the reactions $^{12}\text{C}$, $^{14}\text{Au}$, and $^{16}\text{Pt}$ as shown in Figs. 6, 7, and 8. We also produced it in $^{12}\text{Tl}$ and $^{16}\text{Au}$ experiments. Our redetermination of the $\alpha$-particle energy is 6.260±0.003 MeV.

From an analysis of many decay curves of the 6.260-MeV $\alpha$ group we found a 2-component mixture with half-life periods of 6.5±1 minutes and 1.8±0.5 minutes. The longer lived component corresponds to the $^{206}\text{Em}$ reported by Stoner and Hyde.

The 1.8 - minute period can be assigned to the previously unknown $^{205}\text{Em}$ from arguments based on our excitation function results.

In Fig. 10 we notice that the excitation maximum for the 6.260 MeV peak falls at a higher beam energy than that of $^{207}\text{Em}$. The $^{207}\text{Em}$ peak is distorted
because the Coulombic barrier occurs at a $^{14}\text{N}$ energy close to the true $^{207}\text{Em}$ maximum. Rough decay curves were taken on the 6.260 MeV activity produced at 3 points on the excitation peak, and these indicated that the 1.8 minute half life belonged to $^{205}\text{Em}$ and the 6 minute half life to $^{206}\text{Em}$. These two isotopes decay by electron capture to $^{205}\text{At}$ and $^{206}\text{At}$, respectively. The excitation functions for $^{205}\text{At}$ and $^{206}\text{At}$ are shown in Fig. 11(a). These curves are distorted toward lower energy owing to an experimental difficulty connected with the 30 minute half lives of these isotopes. During the course of the experiments there was a pause of only 4 minutes between counting periods when the absorber foil stack was increased by one. Hence the counting chamber was contaminated with $^{205}\text{At}$ and $^{206}\text{At}$ produced during the previous one or two counting periods. If this distortion is taken into account the $^{205}\text{At}$ and $^{206}\text{At}$ activity can be associated with the 1.8 minute and 6 minute emanation activities at 6.260 MeV. The presence of 5.580 MeV alpha particles of $^{202}\text{Po}$ also supports the $^{206}\text{Em}$ assignment.

In other experiments done with $^{14}\text{N}$ ions slowed by 12, 15, 18, and 19 absorbers, respectively, at times when the emanation counting chamber was uncontaminated with residual astatine activity we measured the ratio of 5.896 MeV $^{205}\text{At}$ activity to 6.260 MeV alpha particles. This ratio changed successively through the values 0.33, 0.24, 0.08, and 0.05. This confirms the fact that $^{205}\text{Em}$ is most prominent on the high beam-energy side of the excitation maximum of the 6.260 MeV activity.

A slight difference in the $\alpha$ energies of $^{205}\text{Em}$ and $^{206}\text{Em}$ was established by making half life determinations on the front and back edges of the 6.260 MeV $\alpha$-peak. The low-energy side was richer in the 6.5 minute component and the high-energy side was richer in the 1.8 minute component, which led us to the following adjusted energy assignments: $^{206}\text{Em}$ 6.258±0.003 MeV and $^{205}\text{Em}$ 6.262±0.003 MeV.
F. Emanation-204

An $\alpha$ activity at 6.416 MeV with a half life of 75±2 seconds was prominent in the emanation fraction from gold targets bombarded with $^{14}$N or $^{16}$O nuclei (Fig. 7) or from platinum targets bombarded with $^{16}$O nuclei (Fig. 8). The excitation function experiments with a thin gold target bombarded with $^{14}$N (Fig. 10) show clearly that the yield of the 6.416-MeV $\alpha$ activity occurs at a beam energy intermediate between those for the maximum yield of $^{205}$Em and $^{203}$Em. This fact and the agreement of the $\alpha$ energy with the approximate value to be expected from systematic trends in $\alpha$-decay energies confirm the assignment of the new activity to $^{204}$Em.

Further evidence for this assignment is the appearance of the expected daughter activities in the samples containing the new activity. Alpha groups of 5.860 MeV and 5.948 MeV were seen, as for example in Fig. 9. The first peak decays with a half life of about 12 minutes and can be assigned to $^{200}$Po (Table II). The half life of the 5.948-MeV peak is about 9 minutes and it can be assigned to $^{204}$At, the electron capture daughter of $^{204}$Em. The excitation curves for these products are shown in Fig. 11(b) together with that of $^{204}$Em.

In order to establish quantitative intensity ratios between $^{204}$Em and its daughters some auxiliary measurements were made in a special counting chamber in which the emanation activity was condensed on a spherical surface cooled by liquid nitrogen. In this apparatus the parent and daughter activities were detected with the same efficiency. Astatine-204 is known to decay 4.5 percent through $\alpha$-particle emission and 95.5 percent through orbital electron capture. From these figures and from the ratio of the $^{204}$At and $^{204}$Em peaks in our spectra we estimate that $^{204}$Em decays 42±10 percent through $\alpha$-particle emission and 58±10 percent through
electron capture. From the ratio of the $^{200}$Po peak to that of $^{204}$Em the $\alpha$ branching of $^{200}$Po was estimated to be $14 \pm 5$ percent. This agrees with the value 12.2 percent reported by Brun, LeBeyec, and Lefort.11

In the present experiments no evidence was found for a 3-min 6.28-MeV $\alpha$ group previously but tentatively assigned $^4$ to $^{204}$Em.

G. Emanation-203 and Emanation-203m

In the emanation fractions from the Hg+C, Pt+O, and Au+N experiments a small $\alpha$ peak was observed at 6.497 MeV and a more intense peak at 6.547 MeV (Figs. 6, 7, and 8). In a series of $\alpha$ spectra taken at 30-second intervals on a gaseous sample removed from a thick gold target half lives of $45 \pm 5$ seconds and $28 \pm 2$ seconds, respectively, were found for these peaks. The same half lives were obtained from samples made by the glow discharge method.

In the excitation function experiments performed with thin gold targets it was found that the yield curves of the two activities were similar (Fig. 10). The maximum yields were observed at 123 MeV $^{14}$N beam energy, which is 17 MeV higher than that corresponding to the peak yield for the 6.416 MeV $\alpha$ group of $^{204}$Em. This suggests an assignment of mass number 203 to the two new activities. Further evidence for this came from the observation of polonium and astatine daughter activities. A peak at 6.060 MeV with a half life of about 4 minutes was ascribed to $^{199m}$Po (Table II). Another peak with a half life of about 7 minutes at 6.085 MeV was identified with $^{203}$At. The excitation curves of the 6.497 MeV and 6.547 MeV emanation activities are very similar to the excitation curve of the sum of the $^{199m}$Po and $^{203}$At peaks (Fig. 11(c)) and dissimilar from all other polonium and astatine curves.
We assign the 45-second 6.497 MeV activity to the ground state of $^{203}_{\text{Em}}$ and the 28-second 6.547 MeV activity to an isomeric state as this choice fits best in the energy versus mass number curve (Fig. 12). More information is required to determine the exact location (excitation energy) of the isomeric state above the ground level.

H. Emanation-202

A short lived $\alpha$ peak with 6.636-MeV energy was observed in the irradiation of gold targets with $^{14}$N and $^{16}$O and of platinum with $^{16}$O (Figs. 7 and 8). By examination of several spectra taken at 15-second intervals the half life was determined to be $13\pm2$ seconds. In the thin target experiments the peak yield of this activity occurred when the energy of the $^{14}$N beam was 140 MeV. The excitation function (Fig. 10) leads to a mass assignment of 202. This assignment is confirmed by the similarity in the excitation functions of the 13-second emanation activity and the 6.18 MeV $^{198}$Po peak, the 5.580 MeV $^{202}$Po peak and the sum curve of the 6.18 MeV $^{198}$Po and the 6.133 and 6.227 MeV $^{202}$At peaks as shown in Fig. 11(d). These polonium and astatine nuclides are the expected daughters of $^{202}_{\text{Em}}$. On the other hand the excitation curve of the 6.636 MeV emanation is dissimilar from all other astatine and polonium excitation curves. This assignment fits well on the mass-energy curve of emanation (Fig. 12).
I. **Emanation-201**

In the spectra taken on emanation fractions from gold and platinum targets bombarded with the maximum energy \(^{14}\text{N}\) or \(^{16}\text{O}\) ions we found alpha groups at 6.703, 6.725, 6.768, and 6.845 MeV. These peaks can be seen in Figs. 7 and 8. Some or all of them must belong to emanation isotopes or isomers below mass number 202. The yield of these \(\alpha\) particles, even at the maximum beam energy, was very low compared to the \(\alpha\) groups of \(^{202}\text{Em}\).

The most prominent of the groups, at 6.768 MeV, had a half life of 3±1 seconds. We tentatively assign it to \(^{201}\text{Em}\) on the following incomplete evidence. The excitation function of the 6.768 MeV peak in Fig. 10 indicates a threshold at a \(^{14}\text{N}\) bombarding energy of 135 MeV corresponding to the maximum in the yield curve for \(^{202}\text{Em}\). Since the threshold energy for excitation of each emanation isotope occurs near the bombarding energy corresponding to the yield maximum of the isotope of next higher mass, this observation argues for a 201 assignment. In addition, a prominent alpha peak at 6.312 MeV, which can be assigned to the daughter product \(^{201}\text{At}\), appears in the same spectra. There was also observed a smaller number of 6.38 MeV \(\alpha\) particles which might be produced by the decay of \(^{197}\text{mPo}\), the \(\alpha\)-decay daughter of \(^{201}\text{Em}\) (Fig. 9(a)).

One objection to the assignment of the 6.768 MeV \(\alpha\) group to \(^{201}\text{Em}\) is that the energy is somewhat high (perhaps by 50 keV) on the basis of our extrapolation to lower mass number of the plot of \(\alpha\)-particle energy versus mass number for the element, emanation (Fig. 12). We suggest that isomerism may occur in \(^{201}\text{Em}\) as well as in \(^{203}\text{Em}\) and that the 6.768 MeV \(\alpha\) group belongs to an isomeric form of \(^{201}\text{Em}\). One of the other groups at 6.703 or 6.725 MeV could possibly be the ground state \(\alpha\) group, but we observed too few particles at these energies to permit us to draw any definite conclusions.
The 6.845 MeV group has a half life of a little less than one second. We assign it tentatively to $^{200}{\text{Em}}$ as this fits on the extrapolated mass-energy curve of emanation (Fig. 12), but we have no clear evidence for this assignment.

IV. DISCUSSION

A summary of our emanation results is given in Table III. The observed alpha particle energies of the neutron deficient isotopes of emanation are shown in Fig. 12 as a function of the neutron number. This figure also includes alpha particle energies for the elements polonium, astatine, francium, radium, and actinium. It can be seen that the trend in alpha energy with neutron number is remarkably similar for these elements, even to details of the step-like shape of the curves.

In the previous discussion of our experimental data, we have presented evidence for isomerism in $^{203}{\text{Em}}$ and possibly in $^{201}{\text{Em}}$. It is reasonable to expect such isomers to exist. Below the 126 neutron shell the lowest lying levels are expected to be the shell model states $p_{1/2}$, $f_{5/2}$, and $i_{13/2}$. A series of isomeric states is present in the odd mass isotopes of the even elements lead, mercury, and platinum (Fig. 13). These states have been given a $13/2^+$ assignment, and they decay with an $M^4$ transition from the $13/2^+$ to a $5/2^-$ level. One may reasonably expect the occurrence of the same type of isomerism in the polonium and emanation isotopes. The principal difference is that in these elements alpha decay can be a competitive and probably dominant mode of decay for the isomeric state. We have placed four odd mass isotopes of polonium on Fig. 12. The evidence for isomerism in these cases comes from the work of Hoff, et al., Siivola, Brun, et al., Tielsch, and of Treytl and Valli.
There are no theoretical calculations of the levels in the light isotopes of emanation and polonium, but some calculations have been made by Kisslinger and Sorensen\textsuperscript{16,17} of the level systems for lead, mercury, and platinum. These calculations are based on a nuclear model in which residual internucleonic forces are approximated by a pairing force and a long range quadrupole force. This model accounts satisfactorily for the trends in the $p_{1/2}$, $f_{5/2}$, and $i_{13/2}$ level energies and in particular for the steady decrease in the energy of the $i_{13/2}$ level as the neutron number decreases.

If this trend should occur also in the polonium and emanation isotopes it could explain why isomeric states with half lives of seconds or minutes are observed in the odd mass isotopes with neutron numbers 111 to 117 and not in isotopes with mass number 125, 123, and 121.

ACKNOWLEDGMENTS

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REFERENCES AND FOOTNOTES

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Table I. Previously reported emanation isotopes with 126 or fewer neutrons.\textsuperscript{1-5}

<table>
<thead>
<tr>
<th>isotope</th>
<th>half life</th>
<th>mode of decay</th>
<th>$\alpha$-particle energy (MeV)</th>
</tr>
</thead>
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<tr>
<td>$^{212}_{\text{Em}}$</td>
<td>23 min</td>
<td>$\alpha$ (100%)</td>
<td>6.264</td>
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<tr>
<td>$^{211}_{\text{Em}}$</td>
<td>16 hour</td>
<td>$\alpha$ (26%) , $\beta$ (74%)</td>
<td>5.847 (33.5%) , 5.779 (64.5%)</td>
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<td>$^{210}_{\text{Em}}$</td>
<td>2.7 hour</td>
<td>$\alpha$ (96%)</td>
<td>6.037</td>
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<td>$^{209}_{\text{Em}}$</td>
<td>30 min</td>
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<tr>
<td>$^{208}_{\text{Em}}$</td>
<td>23 min</td>
<td>$\alpha$ (20%) , $\beta$ (80%)</td>
<td>6.141</td>
</tr>
<tr>
<td>$^{207}_{\text{Em}}$</td>
<td>11 min</td>
<td>$\alpha$ (4%) , $\beta$ (96%)</td>
<td>6.12</td>
</tr>
<tr>
<td>$^{206}_{\text{Em}}$</td>
<td>6-7 min</td>
<td>$\alpha$ (65%) , $\beta$ (35%)</td>
<td>6.25</td>
</tr>
<tr>
<td>$^{205}_{\text{Em}}$ (?)</td>
<td>3 min</td>
<td>$\alpha$ (2%)</td>
<td>6.28</td>
</tr>
</tbody>
</table>
Table II. Properties of isotopes of astatine and polonium with mass 207 or less used as aids in mass assignment of emanation isotopes.

<table>
<thead>
<tr>
<th>Assignment</th>
<th>α Energy (MeV)</th>
<th>Half Life</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>207(^{207})At</td>
<td>5.752±0.008</td>
<td>1.8 hr</td>
<td>12</td>
</tr>
<tr>
<td>206(^{206})At</td>
<td>5.696±0.008</td>
<td>31.0±1.5 min</td>
<td>12</td>
</tr>
<tr>
<td>205(^{205})At</td>
<td>5.896±0.004</td>
<td>26.2±0.5 min</td>
<td>12</td>
</tr>
<tr>
<td>204(^{204})At</td>
<td>5.948±0.003</td>
<td>9.3±0.2 min</td>
<td>12,15</td>
</tr>
<tr>
<td>203(^{203})At</td>
<td>6.085±0.001</td>
<td>7.4±0.3 min</td>
<td>12,15</td>
</tr>
<tr>
<td>202(^{202})At</td>
<td>6.133±0.002</td>
<td>3.0±0.2 min</td>
<td>12,15</td>
</tr>
<tr>
<td>202(^{202})Po</td>
<td>6.227±0.003</td>
<td>2.6±0.3 min</td>
<td>12,15</td>
</tr>
<tr>
<td>201(^{201})At</td>
<td>6.342±0.003</td>
<td>90±4 sec</td>
<td>12,15</td>
</tr>
<tr>
<td>200(^{200})At</td>
<td>6.412±0.005</td>
<td>42±2 sec</td>
<td>12,15</td>
</tr>
<tr>
<td>200(^{200})At</td>
<td>6.463±0.005</td>
<td>42±2 sec</td>
<td>12,15</td>
</tr>
<tr>
<td>202(^{202})Po</td>
<td>5.578±0.005</td>
<td>45±1.5 min</td>
<td>11,12,14,15</td>
</tr>
<tr>
<td>201(^{201})Po</td>
<td>5.677±0.005</td>
<td>15.1±0.3 min</td>
<td>11,12,14,15</td>
</tr>
<tr>
<td>201(^{201})mPo</td>
<td>5.780±0.005</td>
<td>8.9±0.4 min</td>
<td>11,12,14,15</td>
</tr>
<tr>
<td>200(^{200})Po</td>
<td>5.860±0.003</td>
<td>11.4±0.4 min</td>
<td>11,12,13,14,15</td>
</tr>
<tr>
<td>199(^{199})Po</td>
<td>5.950±0.008</td>
<td>5.0±0.2 min</td>
<td>11,14,15</td>
</tr>
<tr>
<td>199(^{199})mPo</td>
<td>6.053±0.005</td>
<td>4.1±0.1 min</td>
<td>11,13,15</td>
</tr>
<tr>
<td>198(^{198})Po</td>
<td>6.178±0.005</td>
<td>1.75 min</td>
<td>11,13,15</td>
</tr>
<tr>
<td>197(^{197})Po</td>
<td>6.280±0.005</td>
<td>5.4 sec</td>
<td>11,13,15</td>
</tr>
<tr>
<td>197(^{197})mPo</td>
<td>6.378±0.005</td>
<td>25 sec</td>
<td>11,13,15</td>
</tr>
</tbody>
</table>
Table III: Summary of new results on emanation isotopes with mass 207 or less.

<table>
<thead>
<tr>
<th>isotope</th>
<th>half life</th>
<th>α-particle energy</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{207}_{\text{Em}}$</td>
<td>6.5±1 min</td>
<td>6.135±0.003</td>
</tr>
<tr>
<td>$^{206}_{\text{Em}}$</td>
<td>6.258±0.003</td>
<td></td>
</tr>
<tr>
<td>$^{205}_{\text{Em}}$</td>
<td>1.8±0.5 min</td>
<td>6.262±0.003</td>
</tr>
<tr>
<td>$^{204}_{\text{Em}}$</td>
<td>6.416±0.003</td>
<td></td>
</tr>
<tr>
<td>$^{203}_{\text{Em}}$</td>
<td>75±2 sec</td>
<td>6.497±0.005</td>
</tr>
<tr>
<td>$^{203m}_{\text{Em}}$</td>
<td>6.547±0.003</td>
<td></td>
</tr>
<tr>
<td>$^{202}_{\text{Em}}$</td>
<td>45±5 sec</td>
<td>6.636±0.003</td>
</tr>
<tr>
<td>$^{201m}_{\text{Em}}$</td>
<td>3±1.5 sec</td>
<td>6.768±0.005</td>
</tr>
<tr>
<td>(?)</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
FIGURE CAPTIONS

Fig. 1. Apparatus for melting metallic targets and for fixing emanation radioactivity on an aluminum foil by the glow discharge technique.

Fig. 2. Apparatus for melting metallic targets in vacuum and for measuring $\alpha$ particles from gaseous sample with semiconductor detector.

Fig. 3. Vacuum apparatus for bombardment of target with beam of $^{12}$C, $^{14}$N, or $^{16}$O from HILAC. Gaseous $\alpha$ emitters diffuse continuously from the target chamber to a counting chamber similar to that shown in Fig. 2. Aluminum absorber foils were used in excitation function experiments. The target itself is not shown in the figure. It consisted of a thin or thick metal foil, or a briquette of stearate powder.

Fig. 4. Alpha spectrum of emanation fraction from thick thallium target bombarded 45 minutes with 0.3 $\mu$Amp current of 140-MeV $^{14}$N. Sample was prepared by glow discharge technique. The 20-minute measurement time started 60 minutes after the bombardment.

Fig. 5. Alpha spectra taken with emanation sample prepared by Tl + 126-MeV $^{12}$C bombardment and deposited on aluminum foil by glow discharge technique. Sample was prepared 4 hours after bombardment. Singles spectrum was 5 min count 22 hours after bombardment. Astatine-211 and $^{211}$Po daughter activities have grown to equilibrium with $^{211}$Em. Coincidence spectrum shows $\alpha$ particles in coincidence with any $\gamma$ radiation. Data were collected for 7 hours starting 15 hours after bombardment. The alpha detectors used in these measurements had a sensitive area of 100 mm$^2$.

Fig. 6. Alpha spectrum of emanation fraction prepared by bombardment of mercury stearate powder with 120-MeV $^{12}$C in the apparatus sketched in Fig. 3. Beam
intensity was about 12 μAmp. Gaseous sample was counted between beam pulses for 1 hour. Because the valve from counting chamber to pump was open slightly during the measurement the long-lived activities are depressed with respect to short-lived activities in the spectrum.

Fig. 7. Alpha spectrum of emanation fraction prepared by bombardment of a 0.001-inch gold foil with 140 MeV $^{14}$N in the apparatus sketched in Fig. 3. Beam intensity was about 0.6 μAmp. Gaseous sample was counted between beam pulses for 15 minutes.

Fig. 8. Alpha spectrum of emanation fraction prepared by bombardment of a 0.001-inch platinum foil with 160 MeV $^{16}$O in the apparatus sketched in Fig. 3. Beam intensity was 0.7 to 0.8 μAmp. Gaseous sample was counted between beam pulses for 25 minutes. Because a very small leak of gas from counting chamber to pump was allowed during the measurement, the long-lived activities are depressed with respect to short-lived activities in the spectrum.

Fig. 9. Four examples of "thin target" alpha spectra obtained by bombarding a 2.5 mg/cm$^2$ (0.00005 inch) gold target with $^{14}$N ions of different energies in the apparatus shown in Fig. 3. The energy of the beam was regulated by placing different numbers of 0.00025 inch aluminum absorbers in front of the target. Beam intensity was about 1 μA and measuring times were 20-25 minutes. These and other spectra were used to prepare the excitation functions shown in Figs. 10 and 11.

Fig. 10. Excitation functions for light emanation isotopes produced by bombardment of a thin gold target (2.5 mg/cm$^2$) with $^{14}$N ions. Lower scale shows number of 0.00025 inch aluminum absorber foils placed in front of the target. Upper scale shows corresponding beam energy. Same integrated beam was used
in each experiment. The indicated Coulombic barrier is a rough estimate from tangent spheres calculation. A very small leak of gas from the counting system to the pump was allowed during the measurements in order to maintain low enough pressure. Thus, the excitation functions of the long-lived activities are somewhat depressed relative to those of the short-lived activities.

Fig. 11. Excitation functions of $^{206}_{\text{Em}}$ and $^{205}_{\text{Em}}$ (a), $^{204}_{\text{Em}}$ (b), $^{203}_{\text{Em}}$ and $^{203m}_{\text{Em}}$ (c), and $^{202}_{\text{Em}}$ (d) shown together with the excitation functions of their daughter activities produced by bombardment of a thin gold target (2.5 mg/cm$^2$) with $^{14}_{\text{N}}$ ions. The daughters of $^{206}_{\text{Em}}$, $^{205}_{\text{Em}}$, and $^{204}_{\text{Em}}$ have half-lives long enough to cause a shift in the position of the maximum yield since there was only a few minutes interruption between the successive 20-25 minute measurements. The lower scale indicates the number of 0.00025 inch aluminum absorber foils placed in front of the target and the upper scale gives the corresponding $^{14}_{\text{N}}$ ion energy. Vertical scales not the same throughout.

Fig. 12. Alpha particle energy versus neutron number for different elements in the area below 126 neutron shell and above 82 proton shell. The solid circles indicate energies assigned to ground state alpha decay. The crosses indicate alpha energies assigned to isomeric states. The open circles indicate alpha transitions to excited levels at the daughter nuclei (alpha fine structure). The emanation curve at 115 and 114 neutrons is uncertain as discussed in text. The astatine curve does not include the later results of Treytl and Valli on the isotopes with fewer than 118 neutrons.

Fig. 13. Island of isomerism below the 126 neutron shell. All odd neutron isotopes of lead, mercury, and platinum shown in solid squares have isomeric states.
assigned to $^{13/2}$ level. Odd mass isotopes of polonium and emanation for which there is experimental evidence for isomerism are also shown. The spin and parity of these isomers are not known. See text for discussion.
Fig. 1.
Fig. 2.
Emanation to counting chamber

Set of absorber foils

Insulator

Permanent magnet

0.00015-in. Ni foil

Beam in

Target chamber

Fig. 3.
Fig. 4.
Fig. 5.
Hg (C, xn) Em

- 6.260 Em^{206}
- 6.140 Em^{208}
- 6.037 Em^{209}
- 5.890 Em^{210}
- 5.755 Em^{207}

Fig. 6.
Fig. 7.
Fig. 8.
Fig. 9.
Fig. 10.
Fig. 11.
Fig. 12.

Neutron number

Alpha-particle energy (MeV)

Po  Em  Fr  Ac

Ra

110 112 114 116 118 120 122 124 126

MUB11506
Fig. 13.
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