Two New Alpha-Particle Emitting Isotopes of Element 105, ^{261}Ha and ^{262}Ha†

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Two new α-emitting isotopes of element 105 with mass numbers 261 and 262 have been discovered. The 8.93-MeV 1.8-sec ^{261}Ha was produced by the reactions ^{250}Cf(^{15}N, $4n$)^{261}Ha and ^{249}Bk(^{16}O, $4n$)^{261}Ha, and the 8.45-MeV 40-sec ^{262}Ha by the reaction ^{249}Bk(^{18}O, $5n$)^{262}Ha. The isotopic assignments are based on both α-recoil milking and mother-daughter time-correlation measurements. An upper limit of 50% for branching by spontaneous fission was set for ^{261}Ha. A so far unassigned 25-sec spontaneous-fission activity was observed in the bombardment of ^{249}Bk with ^{18}O ions.

The relatively long half-life of 1.6 sec for ^{260}Ha, the discovery of which was reported in an earlier Letter,[1] encouraged us to look for additional α-emitting isotopes of element 105. By bombardment of a ^{250}Cf target with ^{15}N ions and ^{249}Bk targets with both ^{16}O and ^{18}O ions we have produced two new α-particle emitters, one an 8.93-MeV 1.8-sec activity assigned to ^{261}Ha, the other an 8.45-MeV 40-sec activity assigned to ^{262}Ha. Genetic relationships between the new activities and the known nuclides ^{257}Lr and ^{258}Lr have been established by both α-recoil milking and time-correlation measurements.

Both the ^{250}Cf and ^{249}Bk targets were prepared by the molecular-plating method and deposited on an area of 0.18 cm^2. The isotopic composition of the 350-μg/cm^2 ^{250}Cf target was 6.1% ^{249}Cf, 86.5% ^{250}Cf, 6.8% ^{251}Cf, and 0.6% ^{252}Cf. Several berkelium targets of 300–400-μg/cm^2 thickness were used; the amount of ^{249}Cf in them due to decay of 314-day ^{249}Bk after chemical separation did not exceed 10%. The targets were bombarded by ^{15}N, ^{16}O, and ^{18}O ions accelerated by the Berkeley heavy-ion linear accelerator (HILAC). A beam-current level of 4 μA (measured as completely stripped ions) was typically used, and the energy of the particles was adjusted to the desired range by a stack of beryllium foils.

The experiments were carried out with apparatus similar to that described in the literature.[1-3] The reaction recoils from the target were stopped by rapidly flowing He gas in a small chamber next to the target. The recoil atoms were swept out of the target region through a 0.4-mm-diam orifice into a rough vacuum where the He jet deposited them onto the periphery of a vertically mounted wheel. The wheel was periodically rotated to place the collected recoil atoms next to a series of peripherally mounted Au-Si surface-barrier detectors. There were seven detector stations arranged equidistantly at 45° intervals around the wheel. Two movable and two stationary detectors were used at each of the seven locations. While one set of movable detectors was recording α-decay events from atoms on the wheel, the other set and its stationary complement were analyzing the daughter α-particle activities which had recoiled off the wheel into the detectors.

The signals from the 28 detectors were amplified by modular units developed in our laboratory, processed by a PDP-9 computer, and stored on an IBM tape. α-decay events between 6 to 10.5 MeV were stored in the first three quadrants of the 512-channel spectra; the last quadrant was used to store α-decay and spontaneous-fission events in the energy range from 10.5 to 200 MeV. In addition, the position of the wheel and the time of occurrence of each event with an accuracy of 50 msec were recorded.

^{261}Ha

The α-particle spectra displayed in Fig. 1 resulted from bombardments of the ^{250}Cf target with 83-MeV ^{15}N ions. The complex group of peaks at about 8.9 MeV has been assigned to ^{261}Ha and its α-decay daughter, 8.87-MeV 0.6-sec ^{257}Lr.[3] Excluding the contribution of ^{257}Lr produced directly in the bombardment, the whole complex group decays with a half-life of 1.8 ± 0.6 sec, or with the same half-life as the 8.93-MeV group. The 6.54-, 6.65-, and 6.77-MeV francium peaks are due to a small amount of lead impurity in the target. The activites left in the detecting crystals as shown by the α-particle spectra recorded by the detectors in the off-wheel position had 10 counts in the 8.75- to 8.90-MeV region for a 37-μA h experiment with a distribution of 5, 1, 1, and 3 counts recorded in the first four stations. Within the 2-sec shuttle period, the quadrants had 4, 5, 0, and 1 of these counts, respectively. The above distribution is compatible with the α-recoil daughter activity being 8.87-MeV 0.6-sec ^{257}Lr,[3] and hence its precursor being 1.8-sec ^{261}Ha. The ratio of detected mother atoms to detected daughter atoms is 4.0 $\pm$1.4, which agrees with the value $5.0\pm^{1.6}_{0.8}$ calculated by taking into account geometry and time factors. A calculation based on spin-independent ($l=0$) equations of Preston[4] and a radius parameter of 9.45 fm give a hindrance factor of 2 for the 8.93-MeV α transition.

The 8.93-MeV 1.8-sec activity was also produced by bombardments of ^{249}Bk with 92-MeV ^{16}O ions. In a 127-μA h bombardment, 25 counts were recorded in the 8.93-MeV peak and 7 α-recoil events in the 8.75- to 8.9-MeV range by the detectors in the off-wheel position.

Additional evidence for the isotopic assignment

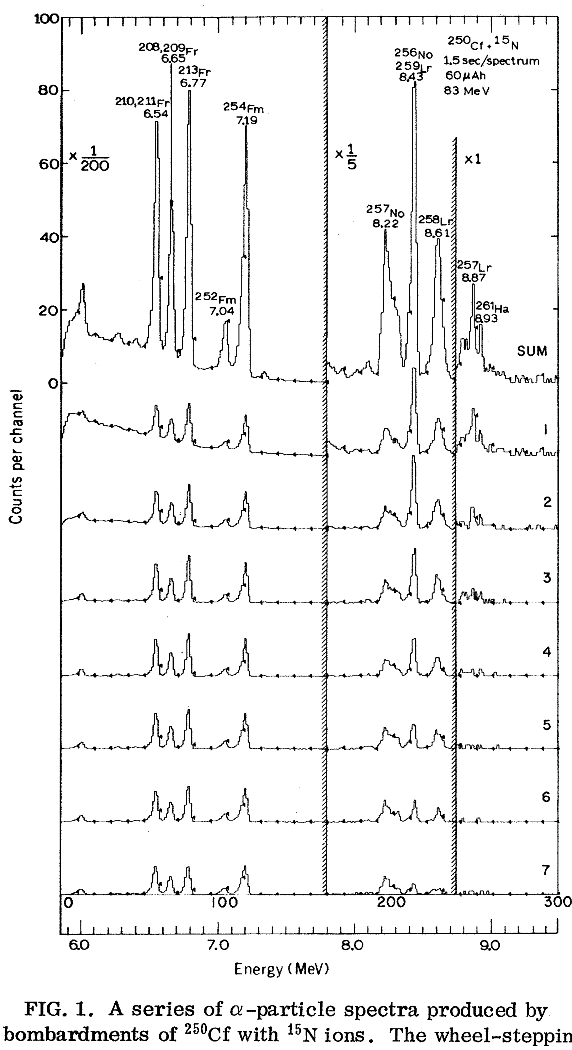

FIG. 1. A series of α-particle spectra produced by bombardments of ^{250}Cf with ^{15}N ions. The wheel-stepping interval, the integrated beam-current reading and the bombardment energy are indicated in the figure. The individual spectra are composed of counts recorded at each of the seven stations by the two movable detectors when facing the wheel. The sum of the seven spectra is plotted in the top curve.

of the new α-particle activity was provided by time-correlation measurements. For each detected α-decay event the α-recoil atom is ejected into the wheel and follows the movement of the wheel until the recoil – or daughter – atom in its turn decays by emitting an α particle. The efficiency of recording the daughter decay is dependent on detector geometry (~35%) and the frequency and duration of wheel and shuttle movements. In order to search for pairs of events due to two successive α decays of the same atom, one specifies the α-particle energy intervals for both events and an upper limit for the time between them. Most random correlations can be eliminated by an analysis of the locations of the detectors which recorded the events and the number of wheel and shuttle motions that took place between them. The distribution of counts recorded in the energy interval of 8.7 to 9.1 MeV for both the mother and the daughter, and following one another within 6 sec, is displayed on the left-hand side of Fig. 2. The solid and open circles mark data obtained from bombardments of ^{249}Bk with ^{16}O ions and ^{250}Cf with ^{15}N ions, respectively. In both bombardments the wheel-stepping interval was 1.5 sec. The α-particle energy for the mother group is 8.93 MeV, while the daughter has a more complex structure with groups at about 8.79 and 8.86 MeV. The half-life of the mother activity is 2 ± 1 sec and that of the daughter 0.8 $\pm^{0.5}_{0.3}$ sec on the basis of the two lower distributions. The properties of the daughter activity are consistent with those of 8.87-MeV 0.6-sec ^{257}Lr[3] and, consequently, the mother is assigned to ^{261}Ha. 13 correlated pairs were recorded in the ^{250}Cf bombardment; the expected number of random correlations is about 0.5 in the whole energy interval of 8.7 to 9.1 MeV.

^{262}Ha

The α-particle spectra displayed in Fig. 3 resulted from a 118-μA h bombardment of a ^{249}Bk target with ^{18}O ions. The new 40 ± 10-sec activity which is assigned to ^{262}Ha has a complex α-particle spectrum with the most prominent peaks at 8.45 and 8.66 MeV. The latter peak is masked by the 8.6-MeV cluster of peaks that belongs to the α-decay daughter ^{258}Lr. The intensity values for the 8.45- and 8.66-MeV α transitions are roughly 80 and 20%; a value of 9.50 fm for the radius parameter yields hindrance factors of 2 and 40 for these transitions. The amount of lead impurity in this particular ^{249}Bk target was very low and therefore the interference from lead-induced activities such as ^{214}At and ^{211m}Po is not serious. The ^{249}Cf present in the target is the source of the ^{256}No and ^{257}No activities in the spectra. The lawrencium activities either are a result of ^{15}N, αxn-type reactions (^{260}Lr and ^{259}Lr partly, at least) or are genetically related to the activities produced in the bombardment (^{258}Lr). The 8.03-MeV 3.0-min ^{260}Lr was first produced in this bombardment.[3]

The α-particle spectra shown in Fig. 4 represent the total of counts recorded by the detectors in the off-wheel position at each of the seven stations. The half-life value for the counts in 8.5- to 8.7-MeV range is 65 ± 25 sec. An examination of the decay of the 8.5- to 8.7-MeV activity within the 8-sec shuttle period gives a half-life of 4.5 ± 2 sec. The ratio of observed mother atoms to recoil daughter atoms is 5.0 ± 1.2 for the 118-μA h bombardment, while the calculated value is 3.1 ± 0.2. Altogether the data are consistent with the parent activity being 40 ± 10-sec ^{262}Ha and the daughter activity 8.6-MeV 4.2-sec ^{258}Lr.

A two-dimensional array of time-correlated events observed in the 118-μA h bombardment of ^{249}Bk with ^{18}O ions is shown in Fig. 5. A maximum of 20 sec was allowed between the occurrence of mother and daughter events. The events in the region enclosed by the broken line stand out from the background caused by random correlations. In the region including the 8.45-MeV peak for the mother and the complex 8.6-MeV peak for the daughter, less than 2 random correlations are expected on the average. The four distributions in the right half of Fig. 2 provide further information about the time-correlated events in ^{249}Bk bombardments with ^{18}O ions. The open circles are the events enclosed by broken line in Fig. 5 and the closed circles mark events recorded in experiments where other ^{249}Bk targets and a 60-sec wheel-stepping interval were used. The α-particle spectrum of the parent activity is complex, with the most prominent peak at 8.45 MeV. Both the time-correlated mother events and the spectra in Fig. 3 indicate that there is an α-particle group at 8.66 MeV. However, as can be seen in Fig. 5 some of the events in this peak seem to be correlated with an 8.50-MeV daughter. The half-life of the mother activity is 43 ± 15 sec, which value is derived from the stationwise distribution of parent events. The daughter activity with a half-life of 5 ± 2 sec and a cluster of α-particle groups centered at 8.6 MeV have the characteristics of 8.6-MeV 4.2-sec ^{256}Lr. The mother is therefore assigned to ^{262}Ha.

No evidence for an activity decaying by spontaneous fission and having a half-life of 2 sec was observed in bombardments of ^{249}Bk with ^{16}O ions or ^{250}Cf with ^{15}N ions. However, because of a background of fission events caused by decay of 2.6-h ^{256}Fm, we can only set an upper limit of 50% to branching by spontaneous fission of ^{261}Ha. In bombardments of ^{249}Bk with ^{18}O ions, a 25 ± 10-sec spontaneous-fission activity was extracted from the ^{256}Fm-induced background as shown by the decay curve plotted in Fig. 6. If this activity is associ-

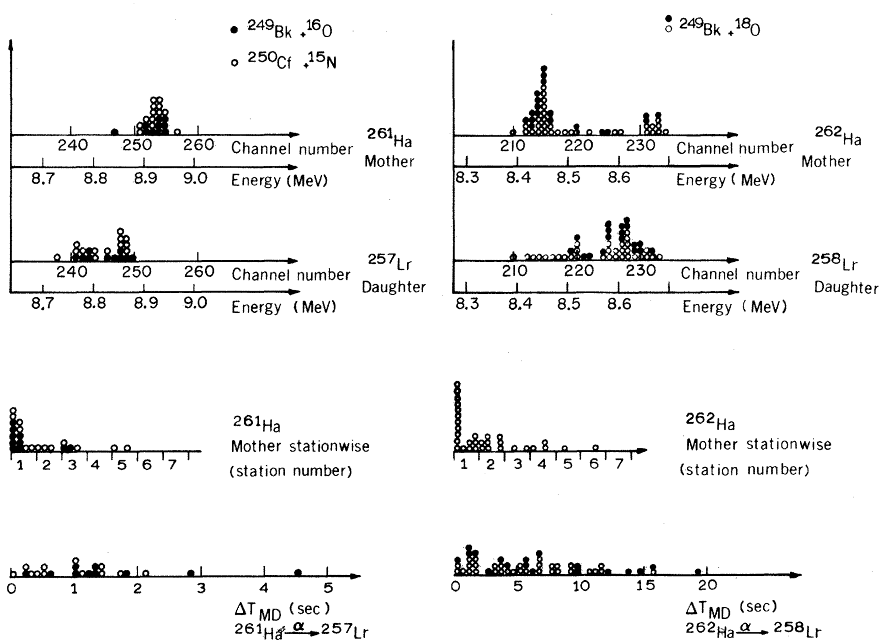

FIG. 2. The distribution of time-correlated events in bombardments of ^{249}Bk with ^{16}O ions and ^{250}Cf with ^{15}N ions (to the left), and in the bombardments of ^{249}Bk with ^{18}O ions (to the right). Energy spectra of mother and daughter events are shown above, the distribution of mother events by station number and quadrant of the wheel-stepping interval is given in the middle, and the difference in time of occurrence between mother and daughter events is displayed at the bottom.

ated with the 8.45-MeV 40-sec α-particle activity assigned to ^{262}Ha, it could arise from branching by spontaneous fission of ^{262}Ha or from its electron-capture decay to ^{262}Rf. On the basis of empirical systematics of spontaneous-fission half-lives,[5] one expects ^{232}Rf to have a half-life of less than a microsecond, and therefore the decay of ^{262}Ha by electron capture followed by prompt decay of the daughter ^{262}Rf by fission would be indistinguishable from spontaneously fissioning ^{262}Ha nucleus in our experiment. Because of the lack of more detailed experimental data, we cannot rule out other alternatives such as ^{263}Ha and ^{263}Rf as possible sources of the 25-sec activity. If the fissions are related to ^{262}Ha, the branching by spontaneous fission or electron capture is about 60%.

Early attempts to identify α-particle-emitting isotopes of element 105 were reported by Flerov *et al.* in 1968.[6,7] Their preliminary conclusion was that they had observed the isotopes ^{261}105 with $E_\alpha = 9.4 \pm 0.1$ MeV, $0.1 < T_{1/2} < 3$ sec, and ^{260}105 with $E_\alpha = 9.7 \pm 0.1$ MeV, $T_{1/2} > 0.01$ sec. In our paper on ^{260}Ha[1] we have discussed these findings in some detail and indicated that they were not compatible with our results. More recently, new work on both spontaneous fission[8,9] as well as α activities[10] assigned to element 105 have been reported by the Dubna group.

In their study of α-particle activities produced by bombarding ^{243}Am with ^{22}Ne ions, Druin *et al.*[10] found evidence for an activity with a half-life of 1.4 ± 0.6 sec and with α-particle groups at 8.9 and 9.1 MeV. The assignment of this activity to either ^{260}105 or ^{261}105 was based on seven time-correlated pairs of events with the mother in the region of 8.8 to 9.2 MeV and the daughter in the region of

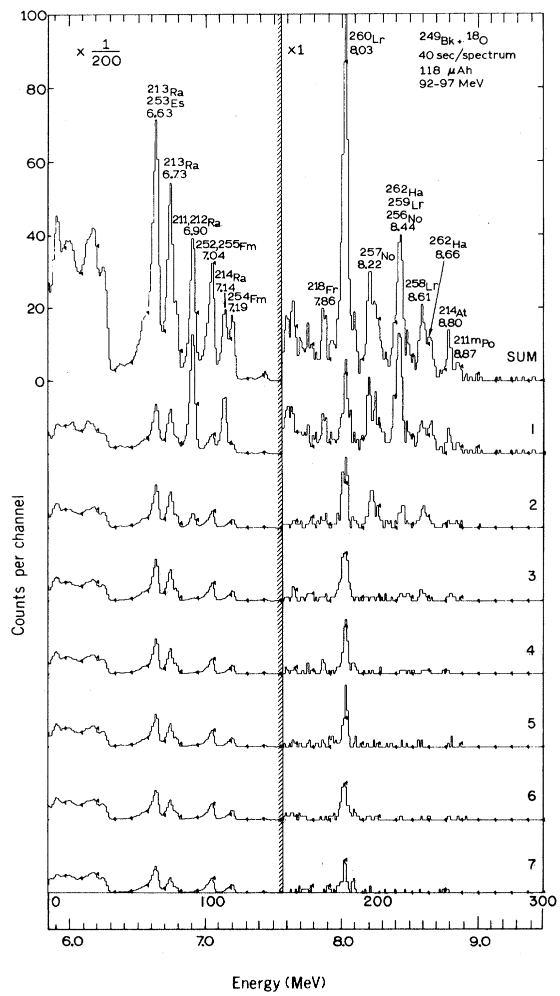

FIG. 3. A series of α-particle spectra resulting from bombarding a ^{249}Bk target with ^{18}O ions. The arrangement of the spectra and the data pertinent to the bombardment correspond to those in Fig. 1.

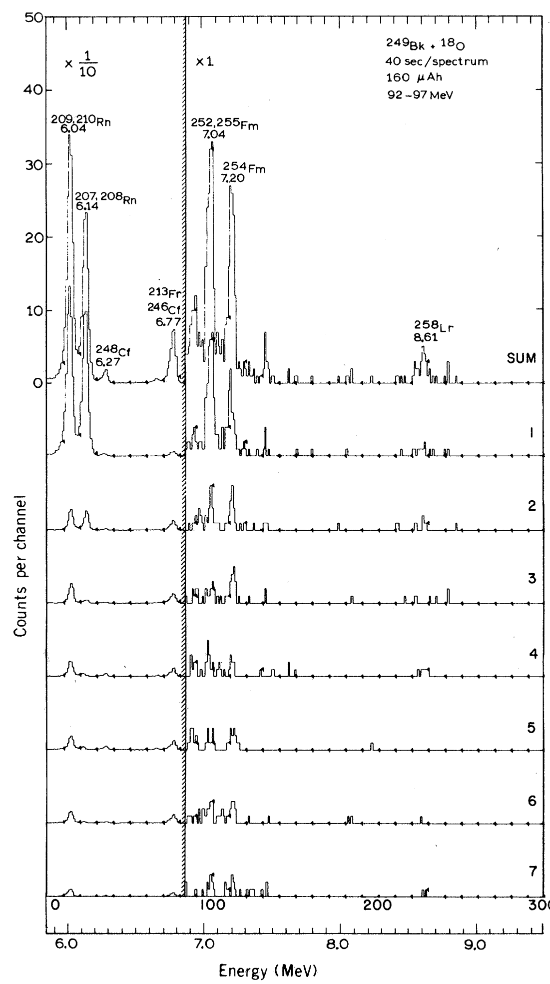

FIG. 4. The α-particle spectra recorded by the detectors in the off-wheel position and resulting from bombardments of ^{249}Bk with ^{18}O ions. The arrangement of the spectra and the data pertinent to the bombardment correspond to those in Fig. 1.

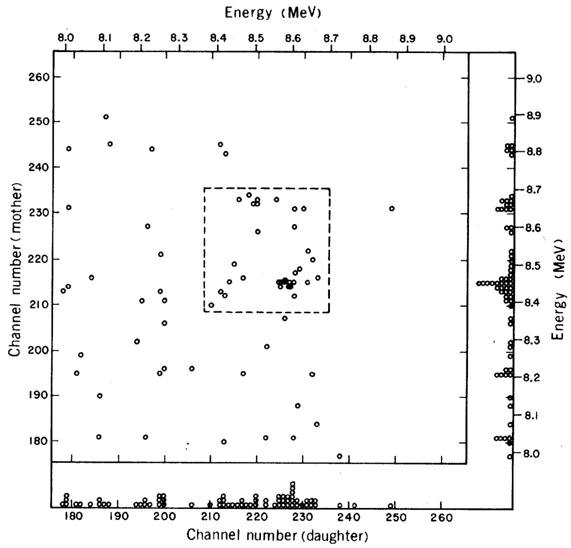

FIG. 5. A two-dimensional presentation of time-correlated events observed in the bombardment of ^{249}Bk with ^{18}O ions. The region enclosed by the broken line was included in the distributions shown in Fig. 2. The histograms on the side are spectra composed of daughter events (horizontal) or mother events (vertical) only.

8.3 to 8.6 MeV. It was assumed that both ^{256}Lr and ^{257}Lr have the same decay properties. Our earlier studies on ^{260}Ha[1] and pertinent Lr isotopes[3] show, however, that this assumption is false and that such correlated events could arise only from the sequence

$$^{260}\mathrm{Ha} \xrightarrow[9.1\ \mathrm{MeV}]{1.6\ \mathrm{sec}} {}^{256}\mathrm{Lr} \xrightarrow[8.3-8.6\ \mathrm{MeV}]{31\ \mathrm{sec}},$$

but not from the sequence

$$^{261}\mathrm{Ha} \xrightarrow[8.93\ \mathrm{MeV}]{1.7\ \mathrm{sec}} {}^{257}\mathrm{Lr} \xrightarrow[8.87\ \mathrm{MeV}]{0.6\ \mathrm{sec}}.$$

Because more detailed information of the energy and time distributions of the correlated events was not given in the paper by Druin *et al.*[10] it is not possible to determine how many of the pairs might actually belong to the former genetic sequence. Moreover, a detailed evaluation of the presented α-particle spectrum is difficult because only a few of the peaks have been assigned to known activities and because the contributions from lead-induced reactions–those producing Th and Pa isotopes in particular–are not discussed.

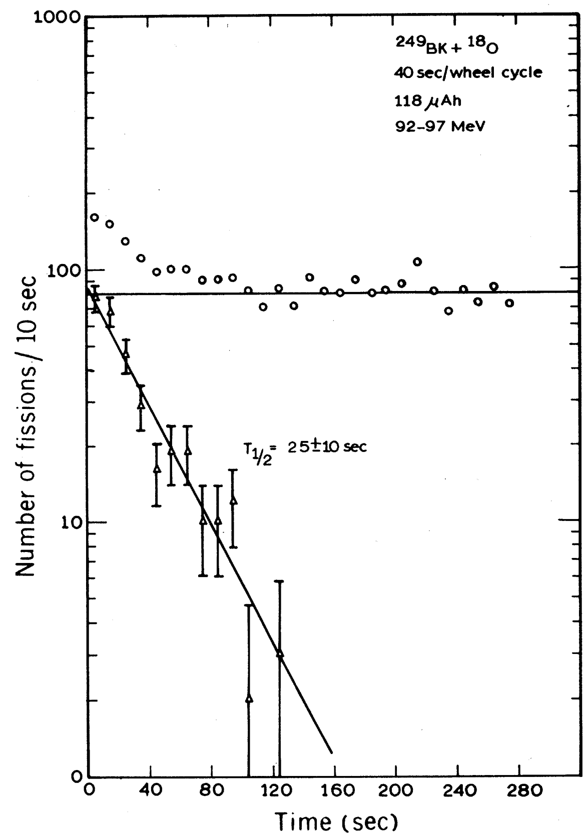

FIG. 6. The time distribution of spontaneous-fission events resulting from the 118-μA h bombardment of ^{249}Bk with ^{18}O ions. The long-lived component is due to ^{256}Fm, the level of which was reduced by starting with fresh spots on the wheel every 80th movement.

The upper limits obtained by us for branching by spontaneous fission for ^{260}Ha,[1] or ^{261}Ha, are so far compatible with the possibility that the 1.8-sec spontaneous-fission activity reported by Flerov *et al.*[8,9] could be due to one of these isotopes.

We wish to express our thanks to the personnel at the HILAC for their contributions in the many phases of this work, and to our colleague James Harris, who was instrumental in the preparation of the targets.

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Compound-Nucleus Contribution to (p, p') Reactions*

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A clean separation is made between the compound-nucleus and direct-reaction contributions to the low-energy continuum region of the energy spectra of protons from inelastic scattering. Bombarding energies are 12 and 17 MeV, and target nuclei are the isotopes of Ni, Zr, Cd, Mo, and Sn. The compound-nucleus contribution is studied by determining level-density parameters and the proton-to-neutron-emission ratios. These are in good agreement with predictions from statistical theory.

INTRODUCTION

The statistical theory of nuclear reactions has been widely used for many years to deduce nuclear level densities from energy spectra of emitted particles, and to calculate cross sections for specific nuclear reactions.[1] In general, results from proton- and neutron-induced reactions have been reasonably consistent and in reasonable agreement with theory,[2] although there have been large inconsistencies with reactions induced by α particles.[3] One persistent difficulty in all of this work has been the question of contributions from direct reactions[2-4]; methods of minimizing it have varied from "plausible" to "highly questionable," but in most cases have left at least something to be desired.

In a recent paper,[5] a method was introduced for achieving a clean separation between compound-nucleus and direct reactions, allowing each to be studied separately and without interference from the other. In this paper, we apply this method to a study of the applicability of statistical theory to compound-nucleus reactions. In a later paper, we shall apply it to studies of direct reactions exciting levels in the high-energy continuum.

EXPERIMENTAL

A block diagram of the electronics used is shown in Fig. 1. Protons of 12- and 17-MeV energy were obtained from the University of Pittsburgh three-stage Van de Graaff accelerator. The targets used in these experiments were self-supporting foils of thicknesses between 1.0 and 3.0 mg/cm^2. The energy spectra of protons scattered from different nuclei were obtained at 75 and 135° laboratory scattering angles. The ΔE-E surface-barrier charged-particle detection telescope used in these experiments consisted of a 50-μ-thick 50-mm^2-area ΔE detector, and a 2000-μ-thick 100-mm^2-area E detector. Particle identification was done with a particle identifier, in the $E\times\Delta E$ mode. Since the energy region of interest is from about 3 to 14 MeV, considerable care was necessary to obtain clean particle identification.

A typical spectrum is shown in Fig. 2. Contamination from carbon and oxygen impurities gives peaks which are easily subtracted off by comparing the spectra with a spectrum from a Mylar target. Background effects become important for high excitation energies where the cross section is small owing to the Coulomb barrier. Back-