First Confirmation of the Discovery of Element 106

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We have confirmed the 1974 report of the discovery of element 106 by Ghiorso *et al.* The 0.9-s $^{263}106$ was produced at the Lawrence Berkeley Laboratory 88-Inch Cyclotron via the $^{249}Cf(^{18}O,4n)^{263}106$ reaction using a beam of 95-MeV ^{18}O ions. Positive identification of $^{263}106$ was made by observing its α decay followed closely in time by the α decay of the ^{259}Rf daughter. Our rotating wheel system was used in a special parent-daughter mode with six pairs of detectors. The $^{263}106$ half-life, α-decay energy, and production cross section are consistent with those measured by Ghiorso *et al.* The half-life and α-decay energies of the ^{259}Rf daughter events are also consistent with those previously published.

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Element 106 was first produced in 1974 by A. Ghiorso *et al.* [1] at the Lawrence Berkeley Laboratory HILAC by bombarding a ^{249}Cf target with an ^{18}O beam to produce the 0.9-s isotope $^{263}106$. The isotope was identified by observing time-correlated α decays from the parent $^{263}106$ and its 3.1-s daughter [2], ^{259}Rf. The decay modes, α-particle energies and half-lives of $^{263}106$ and its daughter activities are shown in Fig. 1. In the 1974 experiments [1], a total of 73 events was observed with α-decay energies near 9.06 and 9.25 MeV, and attributed to the decay of $^{263}106$. Using these events, Ghiorso *et al.* determined the $^{263}106$ half-life to be 0.9 s; 14 of these events were correlated with the 8.770- and 8.865-MeV α particles [2] from the decay of the ^{259}Rf daughter. In addition, the undetected α decay of $^{263}106$ sometimes caused ^{259}Rf daughter atoms to recoil onto movable detectors. These detectors were periodically moved to low-background positions, where the α decays of the ^{259}Rf daughter and ^{255}No granddaughter activities were recorded. The production cross section was estimated to be 0.3 nb.

The confirmation experiment was performed at the Lawrence Berkeley Laboratory 88-Inch Cyclotron. The isotope $^{263}106$ was produced via the same reaction used by Ghiorso *et al.*: $^{249}Cf(^{18}O,4n)^{263}106$. The cyclotron provided a 350 particle-nA beam of 114-MeV (laboratory frame) $^{18}O^{+5}$ which was focused on the californium target contained in the actinide target system. The beam entered through a 2.4-mg/cm^2 Be vacuum window, 0.3 mg/cm^2 of N_2 cooling gas, and the 2.5-mg/cm^2 beryllium target backing before passing through the 0.8-mg/cm^2 californium oxide target material. The target was actually a ^{249}Bk target prepared 16 months earlier, and therefore contained 280 μg/cm^2 ^{249}Bk and 520 μg/cm^2 ^{249}Cf. The beam energy was chosen so that the energy loss in the target cell components resulted in an ^{18}O energy range of 96–94.5 MeV in the californium target, the predicted peak of the $^{263}106$ excitation function [3], and very similar to the energy used by Ghiorso *et al.* [1]. The target thickness was near the recoil range of the compound nucleus reaction products, so they could recoil out of the target into the recoil chamber. The recoil collection chamber, located directly behind the target, was a 1.9-cm-diameter by 10-cm-long cylinder. This volume was continuously swept with He (4.0 l/min, 1.0 atm) containing KCl aerosols to collect the reaction products and transport them through a capillary ($\phi = 2.1$ mm, $l = 7$ m) to the detector array for identification. The transport time of the products from the recoil chamber to the detection site 7.6 m away was approximately 0.4 s.

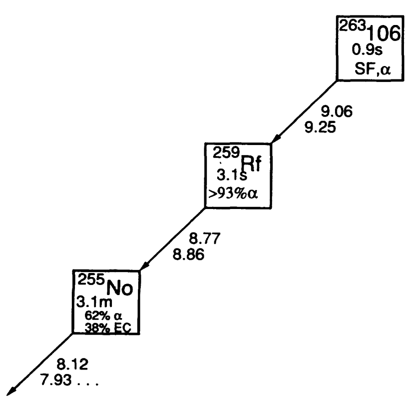

FIG. 1. Decay of $^{263}106$ and its daughter activities.

At the detection site, the MG rotating wheel system and alpha-spontaneous fission detection array [4] was used to characterize the products. The activity from the gas jet was deposited on 40-μg/cm^2 polypropylene foils mounted near the periphery of 20-in-diam fiberglass wheels. A stepping motor was used to turn the wheel and position the foils between six pairs of passivated, ion-implanted planar silicon (PIPS) detectors. Each detector had an active area of 100 mm^2 and a counting efficiency of approximately 30% for alpha particles and 60% for fission fragments. On-line energy calibrations were obtained using the α peaks [5] of 0.5-s $^{211}Po^g$(7.450 MeV), ^{215}Rn (8.674 MeV) in equilibrium with ^{223}Th, and 45-s $^{212}Po^m$(11.65 MeV), all produced from interactions of the ^{18}O beam with the small amount of lead present in the target. The on-line α-particle energy resolution was 60 keV full width at half maximum (FWHM) for the top detectors in the parent mode, and 100 keV FWHM for the bottom detectors in the parent mode. In the daughter

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mode, the energy resolution was 40 keV FWHM.

For this experiment, a special parent-daughter stepping mode was used to facilitate detection of α-α correlations. As shown in Fig. 2, the polypropylene foils were placed in every other hole in the 80-position collection wheel. The stepping was then programmed to proceed in two modes. Figure 2(a) shows the parent search mode in which the activity was collected on the polypropylene foils which were then double-stepped between the first, third, and fifth detector pairs at intervals appropriate for the detection of the 0.9-s ^{263}106. In this mode, the second, fourth, and sixth detector pairs were not detecting activity from sources. If an α particle with the ^{263}106 decay energy (between 8.9 and 9.2 MeV) was observed in a bottom detector, it was assumed that the daughter nucleus, ^{259}Rf, recoiled out of the KCl deposit on the polypropylene foil, and into the top detector. An event within the software gates in the acquisition system generated a signal to initiate the daughter search mode shown in Fig. 2(b) by causing a single step. In this mode the sources were held between detector pairs two, four, and six, with only holes between detector pairs one, three, and five, for a time interval appropriate for the detection of the 3-s ^{259}Rf. Thus, a search for the correlated α decay from ^{259}Rf was initiated in a low background environment without the possibility of random events from the collected sources, while the sources were still being counted in the even detector pairs. At the end of each daughter cycle, the wheel was single-stepped to resume the collection of sources from the gas-jet in the parent search mode.

Many events with energies near that expected for ^{263}106 were detected, causing the initiation of the daughter search mode. Most were due to pileup of lower energy α particles from the decays of 0.84-s ^{8}Li and 0.77-s ^{8}B (Ref. [5]), produced by interactions of the ^{18}O beam with the Be target backing foil. The β decay of ^{8}Li and electron capture of ^{8}B populate excited states in ^{8}Be which then decay into a pair of α particles emitted at 180°. By limiting the ^{18}O beam intensity to 0.35 particle-μA, and limiting the parent and daughter step intervals to 0.6 and 7 s, respectively, the fraction of time spent in the daughter search mode was reduced to 15%. To reduce the acquisition system dead time to $< 1\%$, and to reduce the volume of data collected from this $A=8$ pileup, only events with energies greater than about 7 MeV were recorded. To prevent the recording of noise, data acquisition was disabled for 100 ms during the wheel steps, creating an additional source of dead time.

To prevent the buildup of long-lived activities on the polypropylene foils, the wheel was replaced at 1 h intervals. This also prevented the buildup of large amounts of KCl on the foils, which would degrade the energy resolution in the bottom detectors.

The experiment was run for a total of 60 h of data acquisition time. The spectrum of all α-particle events recorded in the odd-numbered detector pairs during the daughter search intervals is shown in Fig. 3. Only activities which have recoiled onto the top detector surfaces due to the α decay of a parent activity are present. The ^{252}Fm is present as the daughter of ^{256}No. The long-lived activities on the top detectors were characterized in a 30-h post-experiment background measurement. In this measurement, the 7.0-MeV α group was found to decay with the ^{252}Fm half-life of 25.4 h. There was also a small component due to the 6.9-MeV α group of ^{253}Fm, present as the daughter of ^{257}No. ^{211}Po^{g} is present on the top detectors from the α decay of ^{223}Th and its daughter activities. ^{223}Th is produced by the ^{208}Pb(^{18}O,3n) reaction on a small Pb impurity in the ^{249}Cf target. The 7.45-MeV α group of ^{211}Po^{g} decayed with the known 0.52-s half-life for this isotope during the daughter mode intervals. ^{214}Po is present due to the natural content of ^{222}Rn in the air from the decay of ^{nat}U. In the daughter mode spectrum, the region above the ^{214}Po energy is free of activities other than those from the decay of ^{259}Rf and ^{255}No, the daughter and granddaughter of ^{263}106. We observed nine events in the energy range expected for ^{259}Rf and one in the energy range expected for ^{255}No. It should be noted that the events detected in the bottom detectors in the daughter mode have had their energies corrected by 40 keV, since the energy calibrations for the

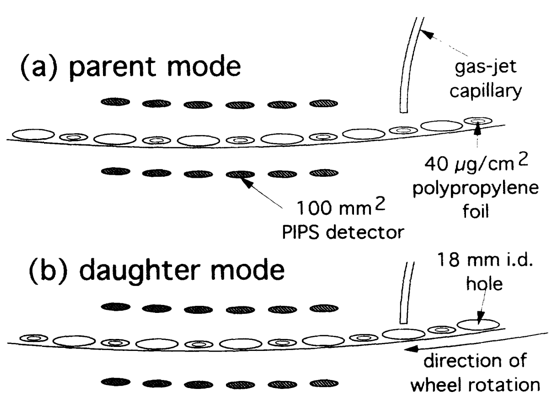

FIG. 2. Schematic of the detection system showing the parent-daughter stepping mode (see text).

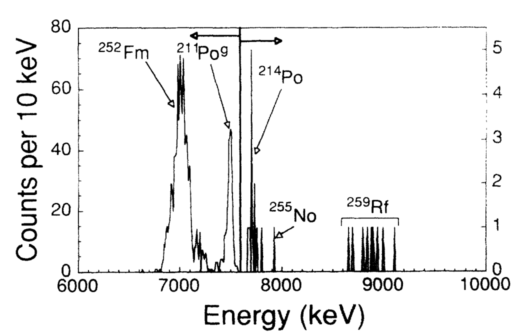

FIG. 3. The spectrum of all α-particle energies recorded in the odd-numbered detector pairs during the daughter search mode. Note the change in vertical scale at 7.6 MeV.

TABLE I. Daughter mode ^{259}Rf events and ^{263}106 events which initiated the daughter mode.

Event #	Detector	Parent Lifetime (s)	Energy (MeV)	Detector	Daughter Lifetime (s)	Energy (MeV)
1				3 bottom	0.717	8.70
2	1 bottom	0.283	8.99	1 bottom	0.717	8.66
3	1 bottom	0.283	9.09	1 top	2.850	8.99
4	3 bottom	1.117	9.00	3 top	1.933	8.93
5	3 bottom	1.183	9.05	3 top	0.267	8.80
6	3 bottom	0.783	9.08	3 top	5.467	8.87
7	1 bottom	0.200	8.95	1 top	10.883	8.90
8	5 bottom	1.733	8.90	5 top	0.517	8.83
9				5 top	4.633	9.10

bottom detectors were performed through polypropylene foils, while in the daughter search mode, there was no intervening polypropylene foil.

Table I lists the detectors, lifetimes, and energies of the nine ^{259}Rf events in the odd detector pairs in the daughter mode. Also listed are the detectors, lifetimes, and energies of the ^{263}106 events that initiated the daughter search mode intervals for each daughter event. In the first and last events, the daughter search mode was initiated by an unrelated event in a different detector pair. Thus, the ^{263}106 parents of these ^{259}Rf events were not detected. If the correlated parent-daughter events were caused by a random correlation mechanism, we would expect only about $\frac{1}{3}$ of the daughter events to be preceded by parent-mode α particles in the same detector station. Instead, each of the first and last events in Table I can be explained in one of three ways:

(1) The ^{263}106 parent α particle was emitted in a direction such that it missed the bottom detector and caused the ^{259}Rf to recoil onto the top detector surface or the top detector holder. The daughter search mode was then initiated by an unrelated event. During this daughter search mode, the ^{259}Rf emitted an α particle which was detected.

(2) The ^{263}106 decayed in the 100-ms interval around the wheel step during which the data acquisition was disabled, causing the ^{259}Rf daughter to recoil onto the top detector surface or the top detector holder. The daughter search mode was initiated by an unrelated event, allowing the decay of the ^{259}Rf daughter to be detected in either the top or bottom detector.

(3) The daughter mode event is due to a long-lived background activity and is unrelated to the decay of any ^{263}106 atom.

A statistical analysis based on the number of α-α correlations observed, detection and recoil efficiencies, half-lives, stepping times, and the numbers of parent and daughter mode intervals resulted in the expectation of 0.24 events of type (1) and 0.25 events of type (2). The observation of one long-lived 8.8-MeV event in the post-run background measurement and assuming this background rate applies to the whole experiment would result in 0.40 events of type (3). The total of expected events of types (1), (2), and (3) is therefore 0.89. The observation of two such events in our experiments is not inconsistent with this value.

The daughter α decay for the seventh event in Table I has a lifetime which is longer than the 7-s daughter interval. This can easily be explained by assuming the ^{259}Rf survived the daughter interval initiated by its ^{263}106 parent. The decay of the ^{259}Rf was then detected during a daughter interval initiated by an unrelated event. A statistical analysis indicates 0.89 of these events is expected, which is consistent with the one event observed.

In the remaining six events in Table I, both the ^{263}106 parent and the ^{259}Rf daughter with normal lifetimes were observed. The lifetimes of the seven parent events were used to determine the ^{263}106 half-life by a maximum likelihood technique [6], resulting in a half life of $1.1^{+2.8}_{-0.6}$ s. The error limits indicate the half-life limits for which the likelihood drops to one half its initial value while holding the initial activity at the value from the best fit. This half-life is consistent with the 0.9 s reported by Ghiorso *et al.* [1]. The distribution of parent energies is fit well with a single α group at 9.01 ± 0.06 MeV after our 100-keV resolution in the bottom detectors during the parent search mode is considered. This is consistent with that of 9.06 ± 0.04 MeV observed by Ghiorso *et al.* [1] for the main α group from the decay of ^{263}106. As a further argument against a random correlation explanation of these data, it should be noted that the energy distribution of these seven parent events differs from that in the overall parent-mode spectra, where the numbers of events decrease sharply with increasing energy.

Similarly, the lifetimes of the eight daughter events were used to determine the half-life of the ^{259}Rf. The 10.883-s lifetime of the seventh event in Table I could not be used since it was outside the daughter mode interval of 7 s, making it difficult to determine a weighting factor for this event. The maximum likelihood fit [6] resulted in a half-life of $1.7^{+0.8}_{-0.5}$ s. This value is consistent with the 3.1 s previously reported [2]. Alpha groups at 8.866 MeV

(40%) and 8.770 MeV (60%) have been reported for the decay of ^{259}Rf (Ref. [2]). The distribution of ^{259}Rf energies in the present work appears to be broader than this, considering the 40-keV resolution in the daughter search mode. There are two reasons for this difference. Since the ^{259}Rf atoms have recoiled into the face of the top detectors, it is possible to detect ^{259}Rf α particles which are emitted in a direction nearly parallel to the detector surface. The detected energy in these events will be lower due to energy loss in the front window of the detector. Also, any conversion electrons which accompany the α particles have a 50% probability of depositing some energy in the detector. This conversion electron energy will sum with the alpha particle energy, producing a prominent high-energy tail on the α peaks. This effect is more prominent than in Ref. [2] because in our experiment the Rf atoms are on the detector surface, resulting in a 50% efficiency for conversion electron summing, and since the depletion depth in our detectors is 300 μm (we assume the detectors in Ref. [2] had 100-μm depletion depth), conversion electrons can deposit more energy in our detectors.

The 7.92-MeV event observed in the daughter spectrum can be interpreted as the decay of ^{255}No present on the detector surface from the decay of ^{259}Rf which had recoiled into the detector from the α decay of ^{263}Rf. A statistical analysis of the number of such events expected based on the number of ^{259}Rf decays observed in the daughter mode, the fraction of time spent in daughter mode, and the 61.4% α-decay branch in ^{255}No results in the expectation of 1.05 daughter-mode ^{255}No events.

The gas-jet yield was determined by measuring the ^{252}Fm produced in this experiment and comparing with production rates calculated from cross sections for binary transfer reactions measured previously [7]. ^{252}Fm production in this experiment was determined by measuring the long-lived α activities in some of the polypropylene foils after removing the wheel from the detector chamber. The gas-jet yield was determined in this way to be near 30% throughout the experiment. Using this gas-jet yield and the other experimental parameters, we calculate the same ^{263}106 production cross section of 0.3 nb reported by Ghiorso *et al.* [1].

We have produced 0.9-s ^{263}106 by the ^{249}Cf(^{18}O,4n) reaction and identified it by correlating the 9.06-MeV α decay of ^{263}106 with the α decay of the 3.1-s ^{259}Rf daughter. The activities were detected on a rotating wheel system which was run in a parent-daughter mode which minimized the effect of random parent-daughter correlations. The measured decay energies and half-lives of ^{263}106 and ^{259}Rf are consistent with those previously published, as is the ^{263}106 production cross section. In our experiment, detection of the 9.06-MeV α particle from the decay of ^{263}106 initiated the low-background daughter search mode, providing a direct correlation between parent and daughter α decays. In the Ghiorso experiment [1], detectors were put into the low-background positions at preset intervals and an indirect correlation between these low-background daughter events and the ^{263}106 parent was made based on a half-life argument.

In 1985 the IUPAP and IUPAC formed a Transfermium Working Group (TWG) to consider questions of priority in the discovery of the transfermium elements. The TWG report [8], published in 1992, assigned credit for the discovery of element 106 to Ghiorso *et al.* based on the experiment described in Ref. [1]. However, in the 1976 article, *Criteria for the Discovery of Chemical Elements* [9], the authors state that, "the name for a new element should not be proposed by the discoverers until the initial discovery is confirmed." Our experiment provides this confirmation of the discovery of element 106.

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