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AN INVESTIGATION OF THE ISOTOPES OF
BERKELIUM AND CALIFORNIUM (thesis)

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OF BERKELIUM AND CALIFORNIUM

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ABSTRACT

The nuclear decay properties of a number of berkelium and californium isotopes synthesized by charged particle bombardments of pile-produced curium and americium isotopes have been investigated. Among the isotopes studied several not previously known were observed. The evidence for their mass assignment is presented. The energies of alpha particles of Bk^{245} and the nuclides assigned as Cm^{245} and Cf^{248} have been measured. Further, the partial alpha half-lives of Bk^{243} and Bk^{245} , the alpha half-lives of the nuclides assigned as Cm^{245} and Cf^{248} , and the partial half-life for spontaneous fission of Cf^{246} have been evaluated. Gamma rays and K x-rays arising from the orbital electron capture of Bk^{243} , Bk^{244} and Bk^{245} and the L x-rays from the decay of Bk^{245} were examined. The orbital electron capture half-lives of Bk^{245} and of the nuclides presumed to be Bk^{244} and Bk^{246} were measured, and a lower limit for the occurrence of this process in Cm^{243} was estimated from experimental data. The assignment of the mass number of the isotope previously referred to as Cf^{246} has been proved.

For the isotopes investigated in this research the observed alpha decay data were found to substantiate the correlations of the alpha

decay systematics. Other questions are discussed, namely the failure to observe Bk^{247} , beta stability in the transcurium region, and lack of evidence for a closed 148 neutron subshell.

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I. INTRODUCTION

The discovery of elements 97 and 98 by Thompson and his co-workers⁽¹⁻³⁾ culminated a long and arduous effort which had as its object not only to observe and identify these new elements, but in addition to systematize extensively many properties of previously known heavy elements in such a way as to enable predictions of nuclear properties to be made which might point the way to a successful attack on other problems of this type. The research reported here includes the further study of the nuclear properties of known isotopes of berkelium and californium and the synthesis of new isotopes of these elements for further investigation. The additional data were needed in order to extend the nuclear decay systematics of the heavy elements, primarily those of alpha decay⁽⁴⁾ and spontaneous fission,^(5,6) and possibly to indicate new correlations. Some of the heavier berkelium and californium isotopes, for example, may possibly possess special stability as a result of the closing of a nuclear subshell.

The problem of producing berkelium and californium isotopes is complicated by the extremely high levels of alpha radioactivity found in the isotopes of curium and americium used as target materials for cyclotron bombardments. Aside from the health hazard created by microgram quantities of americium or curium, which imposes severe restrictions on any operations using these elements, very large separation factors must

be obtained by the chemical procedures. Berkelium and californium were usually produced by bombarding chemically purified americium or curium in a specially designed container. After irradiation the container was opened and most of the chemical separations carried out in a sealed box in which the air was maintained at a pressure slightly less than one atmosphere. The semi-pure samples were removed from the box for further chemical purifications, and the final radiochemically pure samples were examined in the various instruments available for studying nuclear decay properties.

To aid in the understanding of the scope of this investigation a resumé of previous work follows. The isotope of berkelium first discovered was the 4.6 hour Bk^{243} which decays principally by orbital electron capture and exhibits ~ 0.1 percent alpha particle branching.^(1,2) The first isotope of californium was Cf^{244} which emits alpha particles with a 45 minute half-life.⁽³⁾ Later, by a bombardment of U^{238} with C^{12} ions, a 36 hour alpha-emitting californium isotope was found, which was believed to be Cf^{246} .⁽⁷⁾ The chemical behavior of these new elements, studied by experiments with tracer concentrations of the isotopes at the time of their discovery, is that expected of the eighth and ninth members of the actinide series.^(8,9) The most stable oxidation state of berkelium is tripositive, but since it has one electron beyond the half-filled 5f shell it can be oxidized to the plus four oxidation state in aqueous solutions. However, californium has so far been found to exhibit only the tripositive oxidation state in aqueous solutions.

II. EXPERIMENTAL METHODS

A. Pile Production of Target Isotopes

In order to produce new isotopes of berkelium and californium of mass number greater than those previously known it was necessary to produce target material containing larger amounts of heavier isotopes for the cyclotron bombardments. The production of the target material was accomplished by extensive neutron irradiation of certain isotopes of plutonium, americium and curium in chain reactors at other sites. Oxides of the elements irradiated were sealed in metallic capsules, usually aluminum or magnesium. The capsules were especially designed in order to allow water or air cooling, convenient placement or removal from the reactors, and safety provisions for accidental leakage of water in or radioactivity out. From a knowledge of neutron capture and fission cross sections and the neutron flux at the capsule's position it is possible to estimate the relative amount of an isotope that may be produced through a chain of successive neutron captures, even in cases where some of the intermediate isotopes have short half-lives and/or high fission cross sections.⁽¹⁰⁾

The isotope from which most of the heavier americium and curium nuclides are made, Am^{241} , is the beta decay daughter of Pu^{241} , and is removed by chemical separations from the plutonium. The Am^{241} samples were irradiated at the highest possible neutron fluxes for lengths of time required to produce useful quantities of the desired isotopes. A summary of the various isotopes that have been made and the paths of formation are shown in the following schematic diagram.⁽¹¹⁾

If the half-life is not noted in the diagram then it is very long compared to the irradiation time.

One can see that if pure Cm^{244} or Am^{243} is desired then Pu^{242} should be the starting material. Nevertheless, up to the present time an insufficient amount of isotopically pure Cm^{244} or Am^{243} has been synthesized to be profitably used for cyclotron targets.

After the irradiations the capsules were returned to Berkeley for involved chemical separations in which plutonium, americium and curium were individually purified from large amounts of intensely radioactive fission products by combinations of the methods described in Section II-C. The initial chemical separations were usually carried out in enclosed, heavily lead-shielded boxes in which remote control equipment was installed.⁽¹²⁾ Less shielding was necessary as the purification of the transuranic elements progressed. Nevertheless, to contain the large amount of associated alpha radioactivity during any manipulations, the use of a glove box ventilated through a filtered air outlet duct external to the building⁽¹²⁾ was required.

B. Bombardment Techniques

All bombardments were made in the 60-inch cyclotron where a highly developed external target system, the so-called "pistol-grip" type target holder, has been in use for some time.^(2,3) The chemically purified target elements, americium or curium, were evaporated as the nitrates or chlorides in a platinum target cup by induction heating of the platinum. After all the target material had been added and evaporated to dryness the platinum was brought to a red heat to convert the salts to the oxides AmO_2 or Cm_2O_3 .

The beam intensity was continuously monitored by measuring the charge accumulating on the insulated target. The total beam of particles intercepting the target was obtained by recording the current in microamperes. All bombardments in the 60-inch cyclotron were made at maximum particle energy, but the energy of the particles striking the target material was controlled by the thickness of the platinum cup covers.

C. Chemical Separations

In general, the chemical separation procedures were very much the same as those reported earlier,^(2,3,9) but for the sake of completeness a description of them will be repeated.

Following the bombardments the americium or curium oxide was readily dissolved in the platinum target dish with 6 M nitric acid, lanthanum(III) carrier added, and a hydroxide precipitation made with ammonium hydroxide. The hydroxide precipitate could then be redissolved in a smaller volume of solution. The hydroxide precipitate was separated

by centrifugation and dissolved in 3 M hydrochloric acid, after which the solution was made 3 M with hydrofluoric acid. The rare earths and transplutonium fraction, which form insoluble fluorides, were separated at this point by centrifugation from the many fission products which remained in the supernatant solution. The fluoride precipitate was dissolved in 6 M nitric acid, previously saturated with boric acid, which removes the fluoride ion by formation of fluoroboric acid. Another hydroxide precipitation was made with concentrated ammonium hydroxide and the precipitate centrifuged and washed. The hydroxide precipitate was dissolved in a minimum volume of 0.5 M perchloric acid and the solution transferred to the top of a resin bed (18 cm long and 7 to 14 mm diameter) of ammonium form Dowex-50 resin (spherical fines). The column was surrounded by a vapor jacket through which passed trichlorethylene vapor to maintain the temperature at about 87° C. After allowing the radioactivities to be absorbed in a thin band of resin at the top of the column they were selectively eluted with ammonium citrate solution (pH 3.48 - total citrate concentration of 0.25 M) at the rate of 0.03 ml/min/cc of resin bed. The elutriant from the column was collected in small uniform fractions in which the trivalent actinides and rare earths appeared in order of their increasing ionic radius. The californium and berkelium fraction, also containing some of the rare earths eluted in the same fraction, was made acidic (pH ~0 to 0.3) with hydrochloric acid. The activity was adsorbed on Dowex-50 resin and transferred to a column (4 cm long and 3 mm diameter) packed with Dowex-50 hydrogen-form resin. All actinides were eluted

rapidly, in comparison with the lanthanides, with 13 M hydrochloric acid, due to the strong chloride complex formed by the actinides at high chloride ion activity.

In most cases a considerable amount of alpha radioactivity from the americium or curium target material was still present in the berkelium and californium fraction, and had to be separated by several more elutions at elevated temperatures with ammonium citrate from Dowex-50 resin. For the purpose of studying decay properties it is most desirable to have no radioactive impurities in the berkelium or californium fractions, although this goal is difficult to attain when separation factors of 10^{14} are sometimes needed.

D. Counting Equipment and Methods

Thin samples, carrier-free and radiochemically pure, were prepared directly from column elutriant or volatilized from hot tungsten filaments in vacuo. Radioactive solutions, such as column elutriant, were evaporated to dryness upon a platinum disk under a heat lamp. The disk was then flamed to a red heat to burn off organic matter and convert the radioactive elements to inert oxides. Alpha particles were counted in an ionization chamber at a known geometry or examined in the differential alpha particle pulse analyzer which electronically sorts individual alpha particle pulses with respect to voltage (which is proportional to alpha particle energy) and displays the number of counts in each energy increment on a register with a total of 48 increments or channels usable at one time.⁽¹³⁾

Isotopes decaying by electron capture were followed for decay in a methane flow windowless proportional counter which has a high efficiency for detecting ionization produced by Auger electrons. Since the accuracy of several alpha and partial alpha decay half-lives depends on the counting yield in this instrument for the electron capture process, a better approximation than has been used previously was needed. Such data that have been gleaned concerning the counting yield are given in Section III-B. Admittedly it is not to be expected that all electron capture isotopes will have the same counting yield, though the range of percentages found indicates that this approximation is probably good to within a factor of two.

To avoid repeated descriptions of the gamma and x-ray spectrometers and the counting methods used with these instruments, they will be described briefly here and the counters referred to by name in the section on experimental results. Gamma and K x-radiations were studied in a sodium iodide scintillation counter connected to a new modified 50-channel pulse height analyzer.^(14,15) With selected DuMont 6292 photomultipliers and 1 in. x 1 in. cylindrical sodium iodide crystals activated with thallium iodide, about 8-10 percent half-width was obtained for the photoelectric peak from a 660 kev gamma ray. The sodium iodide crystals were surrounded by magnesium oxide used as a diffuse reflector. Energies were determined by direct comparison with pulse analysis of well-known energy standards using the same instrumental conditions. The counting yields for curium K x-rays were determined with the aid of 60 kev photons from the alpha

decay of Am^{241} (16) or with the K x-rays from At^{211} . (17) No corrections of counting yields were made for the counting efficiencies of photons up to 115 kev. From calculations of counting efficiencies for the size of sodium iodide crystal used, (18) essentially all photons entering the crystal with energy less than 120 kev result in the formation of photoelectrons which are counted. The L x-rays of Cm^{245} arising from the electron capture of Bk^{245} were studied in a xenon gas-filled proportional counter (90 percent xenon, 10 percent methane, at 1 atm.) attached to a 10-channel pulse height analyzer. Using the same instrumental and experimental conditions, counting yields and energy calibrations were determined with Np^{237} L x-rays from an Am^{241} source. (16)

III. EXPERIMENTAL RESULTS

A. Berkelium 243 and Bk^{244}

Pure Bk^{243} was produced by the (d,n) reaction from Cm^{242} . An accurate half-life of 4.5 ± 0.1 hours was found which does not differ within the experimental error from the half-life observed of berkelium produced in the early helium ion bombardments of Am^{241} (4.6 ± 0.2 hours^(1,2)). From the alpha disintegration rates of Bk^{243} and its daughter Cm^{243} , determined by alpha pulse analyses, and the known half-life of 35 years for the alpha particle decay of Cm^{243} , (19) the partial alpha half-life of Bk^{243} was calculated to be 130 ± 20 days. The branching ratio for alpha particle emission is ~ 0.15 percent, in contrast with the ~ 0.1 percent found⁽²⁾ for berkelium made from helium ion bombardments of Am^{241} .

There is evidence that leads one to suspect that Bk^{244} is indirectly observed when Am^{241} is bombarded with helium ions. Alpha pulse analysis of Cm^{243} , arising from the electron capture decay of pure Bk^{243} , gave a different abundance ratio between the 5.79 Mev and 5.89 Mev alpha groups⁽²⁾ than the one observed in curium grown from berkelium made by helium ion bombardments of Am^{241} , as one can see in Figure 1. In the americium bombardments the higher abundance of the 5.79 Mev group could arise if Cm^{244} , originating from the orbital electron capture decay of Bk^{244} , were present. It is now known that the energy of the major alpha particle group of Cm^{244} is 5.798 Mev,⁽¹⁹⁾ or nearly the same as found for Cm^{243} . Neither Cm^{243} nor Cm^{244} has detectable alpha particles in the energy range of 5.89 Mev;⁽¹⁹⁾ however, it is possible that the energy of conversion electrons in coincidence with the 5.79 Mev alpha group of Cm^{243} may add to this latter group in the ionization chamber to give an apparent alpha particle peak in smaller abundance at 5.89 Mev.⁽²⁰⁾ Comparison of the curium alpha particle abundance ratios gives a mass yield of Bk^{244} about one-fourth that of Bk^{243} when Am^{241} is bombarded with 33 Mev helium ions. The electron capture half-lives of Bk^{244} and Bk^{243} apparently differ little, if at all. On the other hand, the Bk^{244} partial half-life for alpha decay is probably much longer than that found for Bk^{243} , as can be shown from the lower alpha branching in berkelium produced from Am^{241} .

The gamma and K x-radiations from samples of berkelium produced by 33 Mev helium ion irradiations of Am^{241} , thus containing Bk^{243} and Bk^{244} , were examined in the gamma ray scintillation spectrometer.

All peaks found in the low energy portion of the spectra, illustrated in Figure 2, decayed with a 4.5 hour half-life. An escape peak from the curium K x-rays should have an energy of approximately 85 kev and be in about 20 percent or less abundance;⁽²¹⁾ however, as a more abundant peak was found at 70 kev it can be partially ascribed to a gamma ray. What appears to be gamma radiations with an energy greater than 340 kev is probably Compton spread from higher energy photons associated with the decay of Bk^{243} and/or Bk^{244} as they also decayed with a 4.5 hour half-life.

Limit of electron capture by Cm^{243} .---The energy available for this process calculated from known decay data is 0.005 ± 0.010 Mev. To determine if electron capture takes place with a measurable half-life, a milking experiment planned to discern the growth of Am^{243} from Cm^{243} was attempted. A total of 10 micrograms of curium containing 92 percent Cm^{242} , 5 percent Cm^{243} and 3 percent Cm^{244} (22) was completely freed of all americium by a series of selective elutions from resin columns of Dowex-50 spherical fines operated at room temperature.⁽²³⁾ After allowing the purified curium to decay for 11 months, 10 counts/minute of pure Am^{241} tracer was added to determine the chemical yield of Am^{243} , which was to be separated by another series of resin elutions. Unfortunately, contamination by about 1100 counts/minute of Am^{241} during the later separations made the chemical yield uncertain and reduced the limit of detection

of Am^{243} . With the assumptions of a reasonable chemical yield of 80 percent and that with a thin volatilized sample the limit of detection of Am^{243} would be 0.1 percent of the Am^{241} alpha radioactivity present, a lower limit of 50,000 years was placed on the electron capture half-life of Cm^{243} . A value of 8,300 years was used for the alpha half-life of Am^{243} .⁽²⁴⁾ Of course, the assumptions determine the validity of this limit.

B. Berkelium 245

A new isotope of berkelium decaying principally by orbital electron capture was produced by bombarding approximately 100 micrograms of Cm^{242} , approximately 5 micrograms Cm^{243} and approximately 3 micrograms of Cm^{244} with 33 Mev helium ions and 16 Mev deuterons.⁽²⁵⁾ Examination of the radiations associated with the decay of the berkelium isotopes in the windowless proportional counter showed the 4.5 hour Bk^{243} and a considerably longer-lived decay period corresponding to a 4.95 ± 0.1 day half-life (Figure 3). Some of this radioactivity was examined in the alpha pulse analyzer, which revealed alpha particles of the following energies and abundances (Figure 4): 6.33 ± 0.05 Mev (23 percent), 6.15 ± 0.05 Mev (50 percent), and 5.90 ± 0.05 Mev (27 percent). All three alpha particle groups were observed to decay with a 4.95 day half-life. Initially the three alpha particle groups belonging to the 4.5 hour Bk^{243} were also present. To obtain further confirmation and more data, approximately 180 micrograms of Cm^{242} which contained no more

than 0.06 percent Cm^{243} was bombarded with 33 Mev helium ions, and several targets containing up to 900 micrograms of Am^{241} with 5 to 10 percent Am^{243} present⁽²⁶⁾ were bombarded with 27 to 35 Mev helium ions. In all bombardments the new radiations and half-life were found to be the same as observed in the first bombardment, and in addition a gamma and K x-ray spectrum very similar to that of Bk^{243} and Bk^{244} was observed by use of the sodium iodide scintillation spectrometer. Comparison of Figures 2 and 5 best illustrates the similarity. Photon energies and abundances obtained in the curium bombardment are as follows: 112 kev K x-rays (1.00), 68 kev photon and escape peak of the K x-rays (0.4), and a 245 kev photon (0.3). In the same bombardment the following L x-ray energies and abundances corrected for counting efficiencies were found (Figure 6) by use of the xenon-filled proportional counter: L_{α} - 14.5 kev (1.00), L_{β} - 18.7 kev (1.11), L_{γ} - 21.9 kev (0.35). Assuming Auger conversion factors of 0.1 for K x-rays⁽²⁷⁾ and 0.5 for L x-rays,⁽²⁸⁾ a ratio of K/L orbital electron vacancies of about one can be computed. This number could be readily converted to a K/L capture ratio if the conversion coefficients of the 68 and 245 kev photons in the K and L shells were known. The energies of the K and L x-rays closely approximate those of curium. Comparison of the counting rate of the 4.95 day radioactivity in the windowless counter to that in the 52 percent geometry alpha counter allows calculation of 0.56 percent alpha particle branching, which corresponds to a 24.4 ± 0.8 year partial alpha half-life. Only the statistical error of the counting is given and no error attributed to the 33 percent counting yield of

the proportional counter used, even though this is by far the greatest source of uncertainty. This new berkelium isotope must be of greater mass than 244 or it would have been observed in bombardments of Am^{241} , and it must also have a mass less than 246 as it would be impossible to make Bk^{246} from a helium ion bombardment of Cm^{242} .

Curium 245.—Although Cm^{245} has been identified in mixtures of curium isotopes by means of mass spectrographic analysis,⁽²⁶⁾ the radioactivity associated with this nuclide had not been previously observed. In the work reported here alpha particles of energy 5.34 Mev have been observed in samples resulting from the decay of rather large amounts of Bk^{245} . A target containing nearly a milligram of Cm^{242} and several milligrams of Am^{241} was irradiated with 33 Mev helium ions. Approximately 10 percent of the total curium was Cm^{243} and Cm^{244} and 8.5 percent of the total americium present was Am^{243} .⁽²⁶⁾ In this irradiation sufficient Bk^{245} was produced so that after complete decay of the chemically purified berkelium (the berkelium fraction was separated from curium after the decay of Bk^{243} and Bk^{244}) 0.3 alpha disintegrations per minute with a particle energy of 5.34 ± 0.05 Mev remained in the sample together with Pu^{238} and Cm^{242} impurities (Figure 7). Removal of these alpha radioactive substances left on the sample mounting plate and their elution with Cm^{242} tracer from a cation exchange column with buffered ammonium citrate indicated that only the 5.34 Mev alpha particles were associated with the decay of a curium isotope. Alpha particles with an

energy of 5.34 Mev were also found in a curium fraction chemically separated from pure berkelium after allowing a quantity of Bk^{245} to decay for a short period of time. The ratio of the abundances of the new alpha radioactivity to the amount of Bk^{245} that had decayed remained nearly the same in both instances. The measured alpha particle energy probably does not represent the ground state transition which could be 200 to 300 kev higher in energy by analogy with Cm^{243} . (24) No radiations other than alpha particles could be detected in the separated curium sample. The above evidence indicates a genetic relationship with Bk^{245} , and it seems most likely that the alpha radioactivity originates from a curium isotope with a mass number of 245.

To value the half-life of Cm^{245} it is necessary to know the disintegration rate of the parent, Bk^{245} , during its decay. A quasi disintegration rate is acquired from the K and L orbital electron vacancies, but as mentioned earlier the electron capture rate cannot be found without experimentally measuring the conversion coefficients of the 68 and 245 kev photons in the K and L shells, which is not possible with the instruments available and amount of radioactivity producible. Comparison of the disintegration rates of other isotopes (as determined by the easily measured alpha particle disintegration rate of their daughters) undergoing electron capture to their counting rates in the windowless proportional counter was the basis for assuming a disintegration rate for Bk^{245} . These measurements made

under as nearly identical conditions as possible gave counting yields of 39 percent for At^{211} , 33 percent for U^{231} (29) and 26 percent for Bk^{243} . A value of 33 percent was found for Bk^{245} from the quasi disintegration rate, and this value was used in calculating the half-life of Cm^{245} . Knowing the alpha particle disintegration rate of the Cm^{245} sample and the electron capture half-life of Bk^{245} , about 26,000 years seems to be the best value for the alpha half-life of Cm^{245} .

C. Berkelium 246

It appears likely, but not certain, that some of the expected nuclear decay properties of Bk^{246} have been observed. Berkelium separated from two bombardments of americium (~92 percent Am^{241} and ~8 percent Am^{243} (26)) with 27 Mev helium ions appeared to decay with a half-life that was intermediate between the 4.5 hour Bk^{243} and the 4.95 day Bk^{245} present, as determined by resolving the decay curves. The first bombardment indicated a half-life of approximately one day. In the second experiment the irradiation time and the amount of americium in the target was a factor of three higher than in the first experiment; in this case the half-life resolved from the decay curve (shown in Figure 8) of curium K x-rays detected with the sodium iodide scintillation spectrometer and of the gross radioactivity measured in the windowless proportional counter indicated a half-life of approximately 1.9 days. A disintegration energy of approximately 1.4 Mev is estimated to be available for orbital electron capture by Bk^{246} from energy balances within closed decay cycles.(30) The half-life thought to be associated with Bk^{246} is reasonable for a

highly forbidden transition, which is in order for odd-odd types of nuclei. The mass yield ratio of Bk^{246} to Bk^{245} in the latter bombardment was one-fourth, the same as found for the Bk^{244} to Bk^{243} ratio in the 33 Mev helium ion bombardments of Am^{241} . Different chemical purification procedures were used in the second experiment; these were expected to eliminate any systematic failure to decontaminate the berkelium fraction from fission product radioactivities.

D. Californium 246

In the first bombardment mentioned in the Bk^{245} section, examination of the californium fraction in the alpha pulse analyzer soon after irradiation showed only 6.75 Mev alpha particles decaying with a 36 hour half-life⁽²⁵⁾ (tentatively assigned previously to Cf^{246} .⁽⁷⁾) By use of the alpha pulse analyzer the growth of alpha particles of 6.11 Mev energy was observed and the amount corresponded to a half-life of approximately 160 days. This daughter is undoubtedly Cm^{242} produced by the decay of Cf^{246} , and the assignment to Cf^{246} can be regarded as certain. The decay of the Cf^{246} , followed through a decay factor of approximately 50, gave a half-life of 35.7 ± 0.5 hours in agreement with previous measurements. The ratio of the counting rate in the windowless proportional counter to the alpha counting rate corresponded closely to that observed for a number of "pure" alpha emitters previously examined under similar conditions, so that Cf^{246} appears to be beta stable as expected from consideration of closed energy cycles.⁽³⁰⁾ In this bombardment, no

radioactivity other than Cf^{246} and its daughter Cm^{242} was observed in the californium fraction. This result is somewhat surprising since it might have been expected that Cf^{247} would have a half-life long enough to allow its observation (see Section III-E).

Spontaneous fission rate.⁽³¹⁾--After bombarding curium containing all the isotopes ranging from Cm^{242} to Cm^{245} ⁽²⁶⁾ with 33 Mev helium ions, the californium was separated. By alpha pulse analysis at the beginning of the experiment, the californium fraction was observed to contain essentially pure Cf^{246} radioactivity except for a small fraction of Pu^{238} alpha radioactivity incompletely separated. The amount of Pu^{238} present was insufficient to produce any spontaneous fission events in view of its long half-life for this process.⁽⁵⁾

The fission rate was determined by placing a known quantity of Cf^{246} in a thin deposit on one electrode of a parallel plate ionization chamber, filled with a mixture of argon and carbon dioxide.⁽³²⁾ A stylus recorder maintained a record of each fission event and time of occurrence until essentially all of the Cf^{246} had decayed. The fission rate decreased with the same 35 hour half-life as that observed for the alpha decay of Cf^{246} , thus proving that the fissions recorded originated from this isotope. The experimental data are best summarized in Figure 9. The ratio of the alpha disintegration rate to the spontaneous fission rate is 5.2×10^5 , giving by simple computation the spontaneous fission half-life of $2,100 \pm 300$ years.

E. Californium 248

After the spontaneous fission half-life measurement mentioned above, the remaining Cf^{246} , identified by pulse analysis of alpha particle energy and determination of its half-life, was separated from its daughter Cm^{242} and the californium fraction examined in the alpha pulse analyzer. Alpha particles having an energy different from any of those previously observed for californium isotopes were revealed, and are believed to belong to Cf^{248} produced by the (α, n) reaction from Cm^{245} . The energy measured for these particles is 6.26 ± 0.03 Mev. As can be seen from Figure 10 there was a slight amount of Cm^{242} present in the sample containing the total yield of Cf^{248} . Decay of the alpha radioactivity of this energy followed in the alpha pulse analyzer for seven months indicated that its half-life is 205 ± 40 days (Figure 11), in agreement with the alpha systematics. Correction for residual long-lived alpha radioactivity at 6.26 Mev left after the complete decay of the Cf^{248} will probably decrease the value obtained for the half-life.

In order to determine the spontaneous fission half-life of this nuclide, fission events occurring in the sample were counted by the previously mentioned method. As very few events occurred in the sample of Cf^{248} (about one event every two to five days, which was still much greater than background for such large pulses), the spontaneous fission rate obtained is subject to large statistical variations and errors arising from electronic disturbances; therefore, the value found for the spontaneous fission half-life, 7,000 years, is

rather uncertain. With such poor counting statistics it has not been possible to observe decay of the spontaneous fission rate with the alpha half-life.

It seems very likely that the alpha radioactivity originates from Cf^{248} for the following reasons: from the exponential dependence of the spontaneous fission rate upon Z^2/A of even-even nuclides^(5,6) one can extrapolate from available data⁽³³⁾ that Cf^{248} should have a spontaneous fission half-life of approximately 10^4 years which is in fair agreement, within the limits of error, with the measured value; whereas, from the considerations concerning spontaneous fission described by Seaborg,⁽⁵⁾ an adjacent even-odd isotope of californium would be expected to have a much longer half-life for this process; furthermore, using the alpha half-life found, the bombardment yield is very near the calculated one. The isotope found could not be Cf^{249} because of the absence of target isotope, Cm^{246} , not so far observed; Cf^{247} as a possible assignment seems unlikely as this nuclide is expected to have sufficient disintegration energy available⁽³⁰⁾ to undergo orbital electron capture with a half-life of a day or less, whereas no radioactivity other than alpha particles has ever been detected in the californium fraction in this and a previous similar bombardment where Cf^{247} would have been produced. Nevertheless, the mass assignment is tentative and will remain so until the radiations of Cf^{247} have been observed, or the growth of the daughter Cm^{244} seen.

IV. CONCLUSIONS

A. Remarks Concerning Bk²⁴⁷

In general, no abnormalities have been found by evaluation of the data collected in this investigation. Unusual at the time but later reconciled with the closed energy cycles was the lack of evidence for Bk²⁴⁷. With the assumption of an equal cross section for the formation of Bk²⁴⁷ from the helium ion bombardment of Cm²⁴⁴ as that found for the formation of Bk²⁴⁵ from Cm²⁴², and assuming no other mode of decay, a lower limit of 15,000 years is obtained for the alpha half-life of Bk²⁴⁷. This value is based upon the amount of alpha radioactivity within the energy range of 5.7 to 6.0 Mev. The alpha decay systematics indicate that the alpha particle energy of Bk²⁴⁷ should lie within this range. Very little, if any, Geiger radioactivity remained in the sample after the decay of the lighter berkelium isotopes. If the alpha particle decay is hindered by a factor no larger than that observed for the partial alpha half-lives of Bk²⁴³ and Bk²⁴⁵, then roughly 1,000 years might be expected for the alpha half-life of Bk²⁴⁷. This discrepancy between the lower limit and the value expected might be due to cumulative deviations from the approximations, or more likely, that Bk²⁴⁷ has a short half-life with respect to orbital electron capture. The latter possibility appears more attractive when one considers that the closed energy cycles indicate that approximately 300 kev is available for orbital electron capture,⁽³⁰⁾ and thus a half-life as short as several days might exist. If Bk²⁴⁷ has, in fact, an electron capture half-life

less than 20 days, the detection of its alpha particles and a new decay period in the presence of much larger amounts of Bk²⁴³⁻²⁴⁶ produced in the bombardments described would have been most difficult.

B. Correlation of Data With Alpha Systematics

The exponential relation of alpha decay half-life upon alpha disintegration energy of even-even type nuclei⁽⁴⁾ is obeyed by all the californium nuclides: Cf²⁴⁴, Cf²⁴⁶, and Cf²⁴⁸. On the other hand, using the conventions of reference (4) the alpha particle decay of Cm²⁴⁵ appears to be slightly hindered. The departure factor for the 5.34 Mev group is 5; this factor is about the same as that found for Pu²³⁹. From considerations of the closed energy cycles,⁽³⁰⁾ the energies measured are not those expected for the total disintegration energy, which indicates that the ground state transition is the most highly forbidden. It may be noted that many other alpha particle emitters with an odd nucleon also exhibit a most highly hindered ground state transition. The odd-even berkelium nuclides were found to be further examples, as shown by comparison of the departure factors of the alpha particle groups of Bk²⁴⁵ and Bk²⁴³ listed in the following table.

Nucleus	Alpha energy	Alpha half-life	Departure factor
Bk ²⁴³	0 6.72	430 days	400
	I 6.55	250 days	50
	II 6.20	770 days	5
Bk ²⁴⁵	0 6.33	110 years	1,000
	I 6.15	49 years	60
	II 5.90	90 years	8

In comparison with other known odd-even isotopes (excepting those of bismuth which are near the 82 proton and 126 neutron closed shells), the alpha decay of the berkelium nuclides is very highly hindered. The large hindrance occasioned by the alpha decay across 96 protons might be significant in suggesting special stability at $Z = 96$.

With the assumption that the alpha particle is formed in the nucleus before emission, and that the last odd nucleon is in the highest quantum state, Perlman and colleagues⁽⁴⁾ proposed that pairing of the odd nucleon with a lower lying one already paired will be a slower process than forming an alpha particle from existing paired nucleons. The faster process of forming and emission of alpha particles from nucleons already paired would leave the odd nucleon in an excited state, and the alpha particle emitted would have less than the full disintegration energy available. It is then seen that if the alpha particle is to leave the nucleus with the full kinetic energy available for the disintegration, the odd nucleon must be a component of this alpha particle, and the time required to form such an alpha particle might be much slower than one formed from paired nucleons lying in lower energy levels.

C. Beta Stability

The question of beta stability of many of the isotopes studied is important for the prediction of the nuclear decay properties of heavier isotopes of berkelium and californium and of the isotopes of elements beyond the latter two. The following isotopes have been

found experimentally to be stable or essentially stable toward either emission of negatrons or the capture of orbital electrons: Cm^{243} , Cm^{245} , Cf^{246} and Cf^{248} . No electron capture or negatron decay daughters have been found, nor have any particles or photons not associated with their alpha particle decay been detected during the decay of these isotopes. However, in several cases, such as Cf^{246} and Cf^{248} , the limits of detection have been very poor since insufficient amounts of these isotopes have been synthesized. If one uses the best energy measurements available and estimates alpha disintegration energies for isotopes not now known, quite accurate closed decay cycles involving the above isotopes can be made.⁽³⁰⁾ These closed cycles indicate beta stability for the above mentioned isotopes except Cm^{243} , which appears to be very near stability with respect to electron capture. Although beta stability of Cf^{246} is indicated by the closed cycles, there remains a slight possibility of electron capture because of the uncertainty of the estimates.

D. Spontaneous Fission Rates

It has been noted by Seaborg⁽⁵⁾ and by Whitehouse and Galbraith⁽⁶⁾ that there is a very good exponential dependence of the spontaneous fission rate upon Z^2/A of even-even isotopes. In order to obtain further data to confirm this relation the spontaneous fission half-lives of Cf^{246} and Cf^{248} were measured. For the reasons mentioned in Section III-E the Cf^{248} datum will be disregarded. An extrapolation of previously available data⁽³³⁾ indicated that Cf^{246} , for which Z^2/A

equals 39.04, should have a spontaneous fission "half-life" of approximately 2,000 years. The measured value of 2,100 years is in excellent agreement with that predicted and adds further support to the relation proposed by Seaborg and by Whitehouse and Galbraith.

E. Lack of Evidence for a 148 Neutron Closed Shell

From quantum theory Maria G. Mayer and Haxel, Jensen and Suess have proposed ground state quantum level assignments for nuclear shell structure.^(34,35) It is assumed that the resulting ground state is essentially represented by the angular momentum of the odd nucleon. Large breaks in the energy levels, assumed to be caused by closed shells modified by very strong spin-orbit coupling to produce splitting of levels of high angular momentum, yield the "magic numbers" 2, 8, 20, 50, 82 and 126. A subshell refers to a particular level in a square potential well having the same orbital and total angular momentum ℓ_j . Beyond the 126 neutron closed shell, the theory indicates the filling of the $7i_{11/2}$ and the $6g_{9/2}$ subshells with 22 neutrons to close the $6g_{9/2}$ level at 148 neutrons. One might observe effects due to the closing of a subshell at this point; therefore, an investigation of the properties of such isotopes might yield some knowledge of nuclear structure beyond the 126 neutron closed shell. From the results of this research concerning the spontaneous fission rate of Cf^{246} and the alpha particle energy and half-life of Cf^{248} there seems to be no evidence for a closed subshell at 148 neutrons. When a closed shell has been filled the apparent nuclear radius decreases, and a valley

in the usual smooth trend of the energy surface occurs which means added stability to the nuclides possessing closed neutron or proton shells. An effect of the valley in the energy surface is that for any mode of nuclear decay the disintegration energy is less for decay away from a closed shell and greater for the decay toward it. Closing of a subshell may only have a very small effect on the energy surface and the nuclear radius, particularly if the energy differences between the subshell closed and the next level are quite small, but small irregularities might become apparent when decay properties sensitive to such changes are measured, as, for example, half-lives for alpha and spontaneous fission decay. These means of detection of a filled subshell would depend entirely on the magnitude of the irregularities. On the basis of these considerations one would expect special stability of isotopes containing 138 neutrons, after the complete filling of the $7i_{11/2}$ subshell with 12 neutrons. However, even though this stability is not noticeable, one cannot exclude the existence of a closed subshell at 138 neutrons, but only state that the effects of a closed subshell are so far unobservable. For the same reasons the results reported herein merely illustrate the lack of evidence for a closed subshell at 148 neutrons.

Within experimental errors the measured alpha particle energies and half-lives of all known isotopes containing 148 neutrons, i.e., Cf^{246} , Bk^{245} , Cm^{244} , Am^{243} and Pu^{242} , are not unusual by consideration of the alpha systematics.⁽⁴⁾ The only known isotope decaying to the 148 neutron configuration by means of alpha particle emission,

Cf^{248} , certainly has no higher alpha particle energy or shorter alpha half-life than expected from energy, mass number relations at constant atomic number.

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Fig. 1--Comparison of pulse analyses of alpha particles from curium grown from berkelium produced by (a) bombardment of Cm²⁴² with 16 Mev deuterons and (b) bombardment of Am²⁴¹ with 33 Mev helium ions.

Fig. 2--Gamma rays and K x-rays from berkelium produced by bombardment of Am^{241} with 33 Mev helium ions. The escape peak mainly consists of a gamma ray.

Fig. 3--Decay of berkelium produced by bombarding a mixture of curium isotopes (Cm^{242} to Cm^{244}) with 33 Mev helium ions and 16 Mev deuterons. Measurements were made in a windowless proportional counter.

Fig. 4--Alpha particle pulse analysis of berkelium produced by bombarding a mixture of curium isotopes (Cm^{242} to Cm^{244}) with 33 Mev helium ions and 16 Mev deuterons. The pulse analysis was made after the decay of the 4.5 hour Bk^{243} and Bk^{244} .

Fig. 5--Gamma rays and K x-rays from berkelium produced by bombarding Cm^{242} with 33 Mev helium ions. The measurements were made in the scintillation spectrometer after the decay of the 4.5 hour Bk^{243} and Bk^{244} . Again the escape peak mainly consists of a gamma ray.

Fig. 6--The L x-ray spectrum from berkelium (after the decay of Bk²⁴³ and Bk²⁴⁴) produced by bombarding Cm²⁴² with 33 Mev helium ions. The measurements were made in a xenon filled proportional counter connected to a pulse analyzer. The high counting rate at the lowest energies represents fluorescent L radiation from the platinum mounting plate.

Fig. 7--Alpha particle pulse analysis of the decay products left in sample of berkelium produced by 33 Mev helium ion bombardment of a mixture of curium isotopes (Cm²⁴² to Cm²⁴⁵). Pulse analysis made after the complete decay of Bk²⁴⁵. Curium 242 and Pu²³⁸ appear as impurities in the sample.

Fig. 8--Decay of berkelium K x-rays and gross radioactivity; berkelium obtained by bombardment of Am^{241} (92 percent) and Am^{243} (8 percent) with 27 Mev helium ions. The K x-ray measurements were made by use of the scintillation spectrometer, and the gross radioactive decay followed in the windowless proportional counter.

Fig. 9--Integral decay curve of spontaneous fission events observed in sample of Cf^{246} . T = total number of fissions observed and Δ = number of fissions observed to the time t .

Fig. 10--Comparison of pulse analyses of alpha particles from sample of californium (after decay of Cf^{246}) produced by bombardment of a mixture of curium isotopes (Cm^{242} to Cm^{245}) with 33 Mev helium ions, with energy standards. The pulse analyses were made consecutively using the same instrumental conditions.

Fig. 11--Decay of 6.26 Mev alpha particles originating from californium produced by bombardment of a mixture of curium isotopes (Cm^{242} to Cm^{245}).