

CONFIDENTIAL

UCRL-760
Chemistry-Transuranic Elements

UNIVERSITY OF CALIFORNIA

Radiation Laboratory

Contract No. W-7405-eng-48

THE NEW ELEMENT CALIFORNIUM (ATOMIC NUMBER 98)

S. G. Thompson, K. Street, Jr., A. Ghiorso and G. T. Seaborg

June 19, 1950

CAUTION

This document contains information that is the property of the National Defense of the United States. Its transmission or the disclosure of its contents in any manner to an unauthorized person is prohibited and may result in severe criminal penalties under applicable Federal laws.

Berkeley, California

<u>Standard Distribution: Series A</u>	<u>Copy Numbers</u>
Argonne National Laboratory	1-10
Atomic Energy Commission, Washington	11-12
Brookhaven National Laboratory	13-16
Carbide & Carbon Chemicals Division (K-25 Plant)	17-18
Carbide & Carbon Chemicals Division (Y-12 Plant)	19
General Electric Company, Richland	20-25
Hanford Operations Office	26
Iowa State College	27
Kellex Corporation	28
Knolls Atomic Power Laboratory	29-32
Los Alamos	33-35
Mound Laboratory	36-38
Naval Radiological Defense Laboratory	39
NEPA Project	40
New York Operations Office	41-42
Oak Ridge National Laboratory	43-48
Patent Branch, Washington	49
Technical Information Division, ORE	50-64
UCLA Medical Research Laboratory (Warren)	65
University of California Radiation Laboratory	66-68
University of Rochester	69-70
Total	70

INFORMATION DIVISION
Radiation Laboratory
Univ. of California
Berkeley, California

THE NEW ELEMENT CALIFORNIUM (ATOMIC NUMBER 98)

S. G. Thompson, K. Street, Jr., A. Ghiorso and G. T. Seaborg
Radiation Laboratory and Department of Chemistry
University of California, Berkeley, California
June 19, 1950

ABSTRACT

Definite identification has been made of an isotope of the element with atomic number 98 through the irradiation of Cm^{242} with about 35-Mev helium ions in the Berkeley Crocker Laboratory 60-inch cyclotron. The isotope which has been identified has an observed half-life of about 45 minutes and is thought to have the mass number 244. The observed mode of decay of 98^{244} is through the emission of alpha-particles, with energy of about 7.1 Mev, which agrees with predictions. Other considerations involving the systematics of radioactivity in this region indicate that it should also be unstable toward decay by electron capture.

The chemical separation and identification of the new element was accomplished through the use of ion exchange adsorption methods employing the resin Dowex-50. The element 98 isotope appears in the eka-dysprosium position on elution curves containing berkelium and curium as reference points--that is, it precedes berkelium and curium off the column in like manner that dysprosium precedes terbium and gadolinium. The experiments so far have revealed only the tripositive oxidation state of eka-dysprosium character and suggest either that higher oxidation states are not stable in aqueous solutions or that the rates of oxidation are slow.

The successful identification of so small an amount of an isotope of element 98 was possible only through having made accurate predictions of the chemical and radioactive properties.

THE NEW ELEMENT CALIFORNIUM (ATOMIC NUMBER 98)

S. G. Thompson, K. Street, Jr., A. Ghiorso and G. T. Seaborg
Radiation Laboratory and Department of Chemistry
University of California, Berkeley, California
June 19, 1950

INTRODUCTION

The discovery¹ of element 97 in December, 1949, was immediately followed by a concentration of effort on the search² for element 98. The methods used in the search for these two transuranium elements were similar and the solutions to the most difficult problems in the work on element 97 were directly applicable toward element 98.

All of the experiments completed prior to February, 1950, were unsuccessful for reasons which are now clearly apparent and the present success in this work has been due to relatively recent advances of several types. A very important contribution has been the further development of the systematics of radioactivity,³ particularly in the region of heavy isotopes. The use of these systematics enables fairly reliable predictions of the half-lives and radiation energies of heavy radioactive nuclides to be made. Advances in the knowledge and understanding of the chemical properties⁴ of the heavy elements with improvements in the methods of separation^{5,6} were likewise of great importance in making a solution to the problem

¹Thompson, Ghiorso, and Seaborg, Phys. Rev. 77, 838 (1950).

²Thompson, Street, Jr., Ghiorso, and Seaborg, Phys. Rev. 78, 298 (1950).

³Perlman, Ghiorso, and Seaborg, Phys. Rev. 77, 26 (1950).

⁴G. T. Seaborg, Nucleonics, 5, No. 5, 16 (1949).

⁵K. Street, Jr., and G. T. Seaborg, J. Am. Chem. Soc. 72, 2790 (1950).

⁶Thompson, Cunningham, and Seaborg, J. Am. Chem. Soc. 72, 2798 (1950).

possible. Important contributions were made through the improvement of the methods for the handling and measurement of radioactivity.⁷

In view of the fact that sufficiently intense beams of energetic particles with nuclear charge greater than two have not yet been available, it was clear from the beginning that there were only two methods of approach to the production of synthetic element 98. The first approach, which proved successful, was through the bombardment of curium with helium ions. This approach provided an advantage in that the yields of the (α, xn) reactions were more or less predictable. At the same time it possessed a disadvantage in that the isotopes of element 98 so produced should be neutron deficient and of relatively short half-life as the result of considerable instability towards electron capture and alpha-particle decay. The second approach was through intensive neutron irradiations of curium in order to eventually produce, through successive (n, γ) reactions, curium and berkelium isotopes of masses sufficient to be unstable toward negative beta-particle decay and so produce isotopes of element 98.

This second approach was subject to great handicap in that it involved great uncertainty in the prediction of the mass numbers of the desired curium and berkelium isotopes which decay by beta-particle emission. The first isotope of element 98 so produced might be of mass 248 or 250. Not only is the order of the reaction in which these isotopes are produced very large, but also the cross sections for the (n, γ) reactions cannot be reliably estimated. Thus, in this second approach, the intense irradiation of curium samples with neutrons bears a time scale uncertain within several years. So far this method has been employed without success.

One of the important problems which had to be solved was that of obtaining sufficiently large amounts of curium as sources for the production of element 98. Americium, the source for the production of curium, became available in milligram

⁷Nelson B. Garden and co-workers, unpublished work.

amounts through the neutron irradiation of plutonium.⁸ The curium was produced in smaller amounts by the irradiation of americium with neutrons.^{8,9} Each step required the use of tedious chemical separations.

Since the isotope Cm²⁴² has an alpha-decay half-life of about 160 days, its intense radioactivity presented a major difficulty. The microgram amounts of curium available for the alpha-particle bombardments emit alpha-particles at the rate of approximately 10^{11} disintegrations per minute. This high level of radioactivity necessitated not only the design and development of advanced techniques and equipment for its safe handling⁷ but also made it necessary to attain enormous separation factors of the order of 10^6 in the isolation of an expected 10^3 to 10^4 atoms of the new element from the target material in order to permit the detection of its expected radioactivity, amounting to less than 10^3 disintegrations per minute. Furthermore, this high degree of separation had to be carried out in good yield in a short length of time. In order to provide the greatest sensitivity of detection and thus a possibility of observing the radioactivity of the new element, it was clearly necessary to predict accurately the nuclear properties, including the half-life and radiation characteristics of the isotopes of element 98, so that the experiments could be properly designed. Only then could the optimum period of the bombardment, time of chemical separations and optimum adjustments for the measurement of the radioactivity be established. Likewise, it was necessary to predict the chemical properties of the new element so that its rapid separation from the target material, Cm²⁴², and other isotopes produced by bombardment could be made. An erroneous prediction of any of these properties would have meant unsuccessful experiments.

⁸Ghiorso, James, Morgan, and Seaborg, Phys. Rev. 78, 472 (1950).

⁹Seaborg, James, and Morgan, National Nuclear Energy Series, Plutonium Project Record, Vol. 14B, "The Transuranium Elements: Research Papers," Paper No. 22.1 (McGraw-Hill Book Co., Inc., New York, 1949).

The discovery^{1,10} of element 97 was of primary importance in making possible better predictions of the chemical properties of element 98. In such predictions it was first necessary to consider its normal oxidation state in aqueous solution. Since the oxidation potential of the change $\text{Bk(III)} \longrightarrow \text{Bk(IV)}$ was found to be quite negative^{6,11} (-1.6 volts), it seemed likely that the tripositive state of element 98 would be the state normally existing in aqueous solutions. Since the tripositive oxidation state appeared to be the one on which the chemical separations would have to depend, it was necessary to use the ion exchange methods as they were used in the search for element 97 and to estimate precisely the position of element 98 relative to berkelium and curium in the separations.

Since berkelium had taken its place as the eighth member of the actinide series, there was every reason to believe that element 98 should be an "eka-dysprosium." The properties of berkelium made clear the existence of the same sort of discontinuity in ionic radius at the point of half-filling of the 5f electron shell, as occurs in rare earth elements at the point of half-filling of the 4f shell. Knowing, then, the relation between gadolinium, terbium, and dysprosium on the one hand, and the homologues curium and berkelium on the other, one could estimate with confidence the fractions from the adsorption columns in which tripositive eka-dysprosium (element 98) would be present. Using this information it was predicted that element 98 should be eluted at about 1.4 times the rate of elution of berkelium. Needless to say, element 98 could also be expected to exhibit normal chemical behavior similar to the rare earths--namely, it should carry well with rare earth or actinide fluoride and hydroxide precipitates.

¹⁰Thompson, Ghiorso, and Seaborg, Phys. Rev. (to be published).

¹¹W. M. Latimer, Oxidation Potentials (Prentice Hall, Inc., New York, 1938).

The discovery^{1,6,10} of the isotope Bk²⁴³ and measurement of the energies of its alpha-particles also provided the key to better predictions of the nuclear properties of the isotopes of element 98. By making use of these data to extend the systematics of alpha-radioactivity,³ it was possible to make improved estimates of the alpha-particle energies of the isotopes of element 98 and of other isotopes of element 97. These estimated alpha-particle energies could in turn be used to estimate the alpha-decay half-lives of the transcurium isotopes³ and also to calculate by closed decay cycles the total energies for electron capture or beta-decay. The electron capture disintegration energies then led to the estimation of half-lives for this mode of decay, using an empirical energy versus half-life relationship,¹² which, although very rough due to uncertainties in the degree of prohibition, was nevertheless very useful in attaining the half-life estimates. By means of these methods, it was estimated that the longest-lived isotope of element 98 that could be produced by the bombardment of the isotope Cm²⁴² with helium ions should be the isotope 98²⁴⁴ by the ($\alpha, 2n$) reaction, with an anticipated half-life between 30 minutes and several hours and emitting alpha-particles with energy in the range 7.0 to 7.3 Mev.

The high energies of the alpha-particles emitted in the decay of the isotopes of the new element were expected to make possible a sensitive means for their detection. Such high energy alpha-particles are conveniently observed with great sensitivity and their energies measured by means of a differential alpha-energy pulse analyzer.¹³ In using our instrument in the most sensitive manner, it is necessary to limit the total amount of alpha-radioactivity in the sample examined

¹²S. G. Thompson, Phys. Rev. 76, 319 (1949).

¹³Ghiorso, Jaffey, Robinson, and Weissbourd, National Nuclear Energy Series, Plutonium Project Record, Vol. 14B, "The Transuranium Elements: Research Papers," Paper No. 16.7 (McGraw-Hill Book Co., Inc., New York, 1949).

to less than about 10^4 disintegrations per minute. Thus the enormous amount of curium radioactivity in the target material used for producing element 98 had to be separated from the isotopes of the new element by a factor of some 10^6 to 10^7 , as mentioned previously, before the alpha-energy pulse analyzer could be used.

The observation of alpha-particles of energy 7.0 to 7.3 Mev is of particular significance in itself. Radiations of such energy have been observed previously only in the decay of the isotopes of elements just above lead as members of distinctive chains of alpha-emitters.¹⁴ These isotopes are of so much lower mass that they cannot be produced in bombardments with helium ions of less than 40-Mev energy.

For these reasons the observation of distinctive alpha-particle radiations should be expected to provide good evidence for the existence of element 98. Furthermore, evidence based on the alpha-particle radiations, together with the observation of the chemical properties expected for the element by means of which all of the other elements above and including lead are separated, should constitute proof of the existence of the new element.

The first successful experiment² in which an isotope of element 98 was observed was completed February 9, 1950. This experiment was designed to allow the detection of the long range alpha-particles of an isotope decaying with a half-life as short as 20 minutes.

EXPERIMENTAL WORK

Extraction of Curium

The Cm^{242} used as target material for the production of element 98 was separated from strongly neutron-irradiated americium in which it was produced as a daughter of the beta-particle decay of the 16-hour Am^{242m} . The curium was separated from americium and purified by combinations of precipitation and ion exchange methods.

¹⁴G. T. Seaborg and I. Perlman, Revs. Mod. Phys. 20, 585 (1948).

Preparation and Bombardment of Targets

The curium targets were prepared for bombardment in the Crocker Laboratory 60-inch cyclotron by the evaporation of solutions containing microgram amounts of curium nitrate in small platinum dishes of about 0.5 cm^2 area followed by ignition to form curium oxide. In some of the experiments Am^{241} was present to facilitate handling of the smaller amounts of curium and to produce Bk^{243} for purposes of comparison. These targets were bombarded in the same target assembly and in the same manner as has been described previously in a paper on berkelium.¹⁰ The energies of the helium ions used in the bombardments were about 35 Mev (actually more nearly 33 Mev). The intensity of the bombardments averaged about 3 microamperes per square centimeter of target area and the time of bombardment was usually 2 to 3 hours. The helium ion beam also contained some deuterons of energies approximately one-half the energy of the helium ions. (The presence of the isotope Bk^{243} observed in the experiments where Am^{241} was absent was due to the reaction $\text{Cm}^{242}(\text{d},\text{n})\text{Bk}^{243}$.)

Chemical Procedure

Following the bombardment, the platinum dish containing the sample was removed to a glovebox¹⁰ and the curium oxide dissolved in 6M HNO_3 with heating (about 75°C). Curium hydroxide was precipitated with added lanthanum carrier by the addition of excess ammonium hydroxide and was separated by centrifugation. The hydroxide precipitate was dissolved in dilute perchloric acid (0.5M) and the mixture of radioactivities was adsorbed on a small amount of ammonium form Dowex-50 resin (spherical fines) which was then transferred to an approximately 17-cm length, 2-mm diameter column packed with the same resin. The column was surrounded by a vapor jacket through which passed trichloroethylene vapor to maintain the temperature at about 87°C . The elution was performed by passing ammonium citrate (buffered with citric acid to pH 3.5—total citrate concentration 0.25M) through the column at a rate

of 1 drop (about 0.030 cm^3) about every two minutes and the drops were collected separately or in groups of two in the region where the actinide elements were expected. Element 98 was collected in drops of elutriant from the columns just preceding those containing berkelium.

Each drop or a suitable fraction of each drop collected from the column was evaporated on a platinum plate using a heat lamp. Ignition of the plates to red heat removed the citric acid. Each of these samples which preceded the curium peak was examined for high energy alpha-radioactivity using the differential alpha-energy pulse analyzer.

The fractions which should contain elements 97 and 98 were submitted in some experiments to further separation from remaining radioactive contaminants (mainly approximately 10^4 alpha-disintegrations per minute of Cm^{242}) by means of a second Dowex-50 resin column operated in a manner similar to the first column run. In this case, the appropriate elutriant fractions from the first column containing the ammonium citrate were acidified with hydrochloric acid to make the hydrogen ion concentration about 0.1M . Then the mixture of radioactivities was adsorbed on Dowex-50 resin and the column separation made as described previously. (The separation of curium from element 97 and element 98 in one column run as described above is limited to a factor of approximately 10^6 to 10^7 . After the second column run the curium present in element 98 fractions is reduced to a few alpha-disintegrations per minute.) In some of the experiments the lengths of the resin columns were shortened to about 15 cm length in order to increase the speed of separation. Each drop of elutriant preceding the curium peak was again examined for high energy alpha-particles in the same manner as described above in connection with the first column separation. In addition to differential alpha-energy pulse analysis, certain fractions obtained in some of the experiments were also counted in a windowless proportional counter in order to look for soft electrons with

high efficiency. In this case it was possible to observe the electrons emitted by some of the radioactive fission products such as yttrium, which were produced in the bombardments.

Some of the fractions containing element 98 were used for independent investigation of the chemical properties of the new element. These investigations included separations in a Dowex-50 resin column with concentrated (13M) HCl for elution and experiments employing powerful oxidizing agents with carriers such as zirconium phosphate and lanthanum fluoride.

Experimental Methods Used in Radioactivity Measurements

The thin deposits on platinum of the fractions expected to contain element 98 were examined in the differential alpha-particle pulse analyzer. In this instrument individual pulses from an ionization chamber are sorted electronically and recorded on 48 fast mechanical registers in such a way as to separate the individual alpha-particle energies from a mixture of alpha-emitters. In establishing values for the alpha-particle energies of the new isotope, direct comparison was made with pulse analyses of thin samples of isotopes whose alpha-energies are well known, using the same instrumental conditions.

No attempt was made to observe electrons and electromagnetic radiations in the element 98 fractions because of the possible presence of unseparated fission product isotopes in larger abundances which emit similar radiations.

Results of Radioactivity Measurements

The application of the procedures described above to helium ion bombarded targets of Cm^{242} and the examination in the pulse analyzer of the separated fractions expected to contain element 98 revealed a small amount of 7.1 ± 0.1 Mev alpha-particle radioactivity together with the expected amount of residual 6.08 Mev Cm^{242} alpha-particle activity.

The results of a typical experiment in which the alpha-particle energies and rate of decay were measured by differential pulse analysis are shown in Fig. 1. The rate of decay was measured by plotting the area under the 7.1-Mev energy peak versus the time of decay of the radioactivity. This peak was observed to decay with a half-life of approximately 45 minutes through a decay factor of more than 10, as shown in Fig. 2. The tailing out of this curve is caused by a relatively high "electronic" background from the "piling up" effects in the amplifier of the intense Cm²⁴² activity.

The presence of the 45-minute radioactivity was observed in four separate experiments. It was observed in the presence of a relatively large amount of unseparated Cm²⁴² (approximately 10^4 alpha-disintegrations per minute), upon completion of the operation of the first resin column as shown in Fig. 1. The presence of the isotope Bk²⁴³ produced either by (d,n) or (α,2n) reactions in the separate bombardments was not observed after only one column separation because of the closer proximity of the berkelium elution peak to that of curium and the consequent larger amount of curium activity in the berkelium samples. However, after the second column run in which the residual Cm²⁴² radioactivity was separated, it was possible to observe the presence of the separate elution peaks due to the 45-minute activity, Bk²⁴³ and Cm²⁴² in the alpha-particle pulse analyzer. The electron and electromagnetic radiations of the 4.6-hour Bk²⁴³ were also observed by the methods described in reference 10. The best values for the alpha-particle energies and half-life of the 45-minute activity were obtained on the larger samples resulting from the operation of the first resin column. Although samples from the second column run were essentially free of Cm²⁴² contamination, the 45-minute radioactivity had decayed during this additional time interval by a factor of approximately six so that no advantage was gained by the second separation insofar as the radioactivity measurements were concerned. However, important

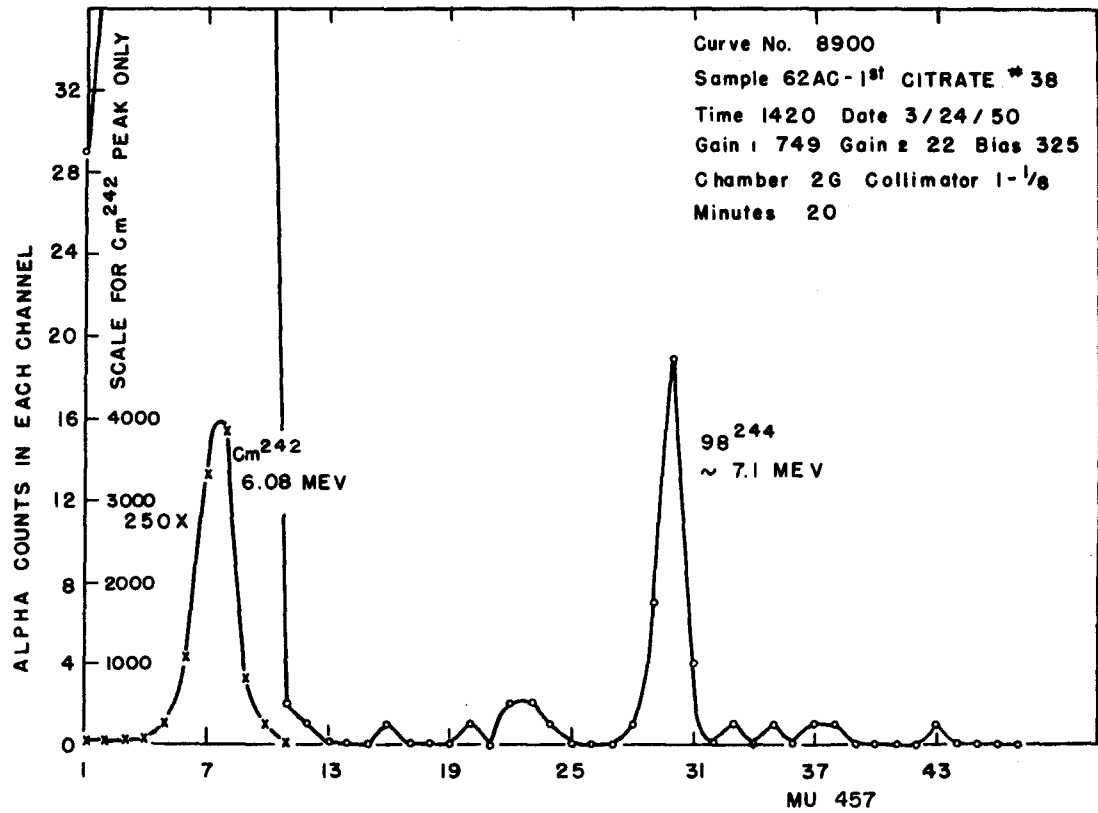


Fig. 1. Alpha-particle pulse analysis on element 98 fraction from first resin column showing 7.1-Mev alpha-particle radioactivity in presence of large amount of Cm²⁴² alpha-radioactivity.

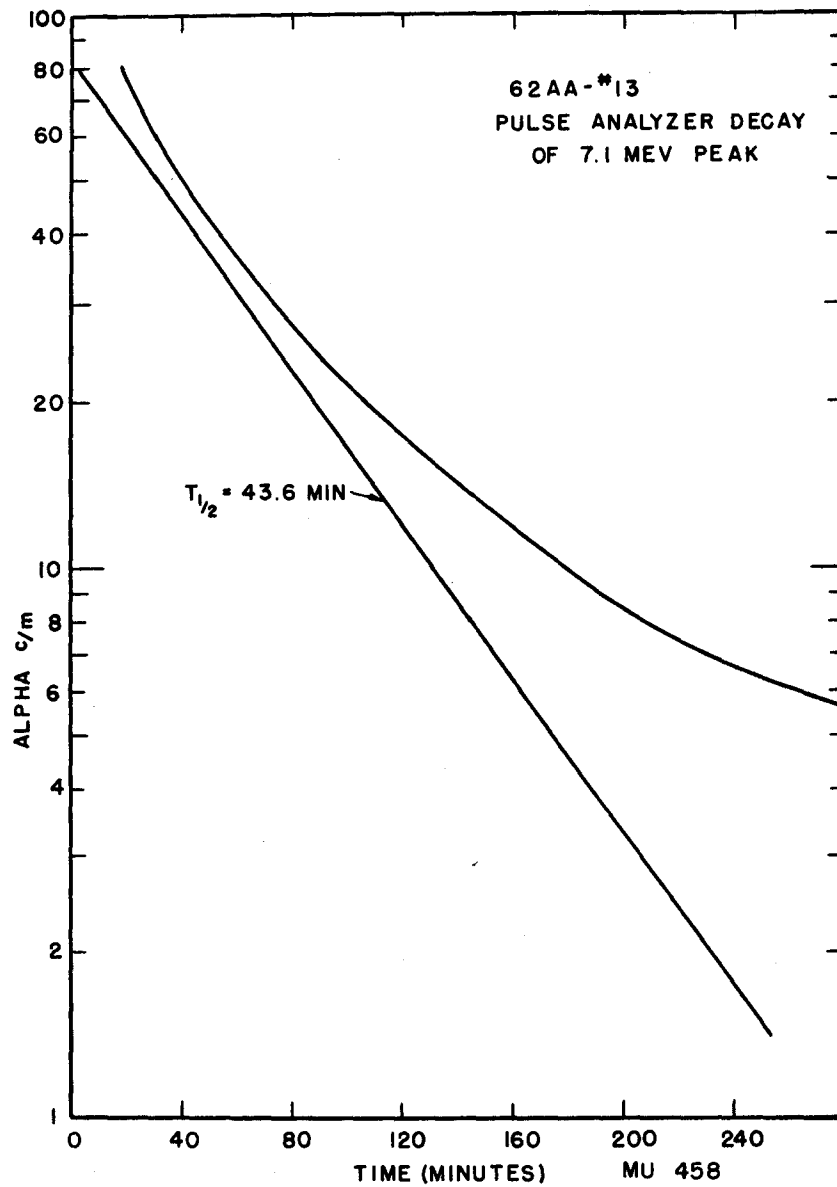


Fig. 2. Decay curve obtained by plotting area under 7.1-Mev peak from pulse analyzer data versus time.

information was obtained concerning the chemical identity, since the isotope Bk²⁴³ was observed to separate from the 45-minute radioactivity.

The Search for Daughters of the Alpha-Particle Decay

The fractions containing the 45-minute activity resulting from separations on the second resin column were examined for possible daughter radioactivities. In these fractions alpha-radioactivity from Cm²⁴² had been reduced to approximately one disintegration per minute. Careful examination of these fractions using the alpha-particle pulse analyzer failed to show any alpha-particle peak other than that of the residual unseparated Cm²⁴² after the 45-minute activity had decayed completely. On the assumption that the 45-minute activity is 98²⁴⁴, the amount of its alpha-radioactivity present in the initial fractions should have produced approximately 0.1 disintegration per minute of alpha-radioactivity due to the alpha-decay daughter Cm²⁴⁰. The level of the background radioactivity in the pulse analyzer chamber was too high to show a definite identification of this amount of daughter isotope.

Cross Section for Formation of Element 98 Isotope

The cross section for the production of the 45-minute activity in the bombardment of Cm²⁴² with approximately 33-Mev helium ions was calculated as roughly 3×10^{-27} cm². In this calculation, decay is assumed to be entirely by the emission of the alpha-particles which were observed. A comparable rate of decay by electron capture is entirely possible and this would of course lead to a proportionately higher cross section. The chemical yield of the 45-minute radioactivity was estimated as approximately 90 percent, based on measured values for separations of the same type. Measured values of the effective weight of target material, the cyclotron beam and time intervals were used in the calculations.

Results of the Chemical Separations

The behavior of the new 45-minute activity in the ion exchange procedures used in its separation are highly significant for the assignment of the atomic number 98. Fig. 3 shows the results of one of the separations in the first resin column after which the Cm^{242} present in the element-98-containing fractions was reduced to approximately 10^5 alpha-disintegrations per minute. The results of one of the separations in the second resin column are shown in Fig. 4. In this case the position of the 45-minute radioactivity is shown relative to berkelium, curium, and yttrium. The alpha-radioactivity was measured in all cases except that of the fission product yttrium, whose beta-radioactivity was measured. These results might be summarized in the statement that element 98 is eluted in the expected eka-dysprosium position preceding berkelium from the resin column.

A region of particular importance is illustrated in Fig. 5, which compares the elution positions of the actinide group element 98-berkelium-curium-ameridium with the homologous lanthanide group dysprosium-terbium-gadolinium-europium. The comparison is based in part on a number of separate experiments performed under as nearly identical conditions as possible. The ordinates are normalized to show equal amounts. These particular results might be summarized as follows: the difference in rates of elution between element 98 and berkelium as compared with the difference between berkelium and curium is remarkably similar to the spacing dysprosium-terbium and terbium-gadolinium. There is a remarkable similarity in the spacing of the four members of each group, with breaks at the points of half-filling of the f electron shells, gadolinium and curium; the spacings here reflect the relative changes in ionic radii which determine the relative separations in the ion exchange adsorption method.

With regard to the group separation of actinides from rare earths using the Dowex-50 resin column with 13M hydrochloric acid,⁵ the new 45-minute activity was

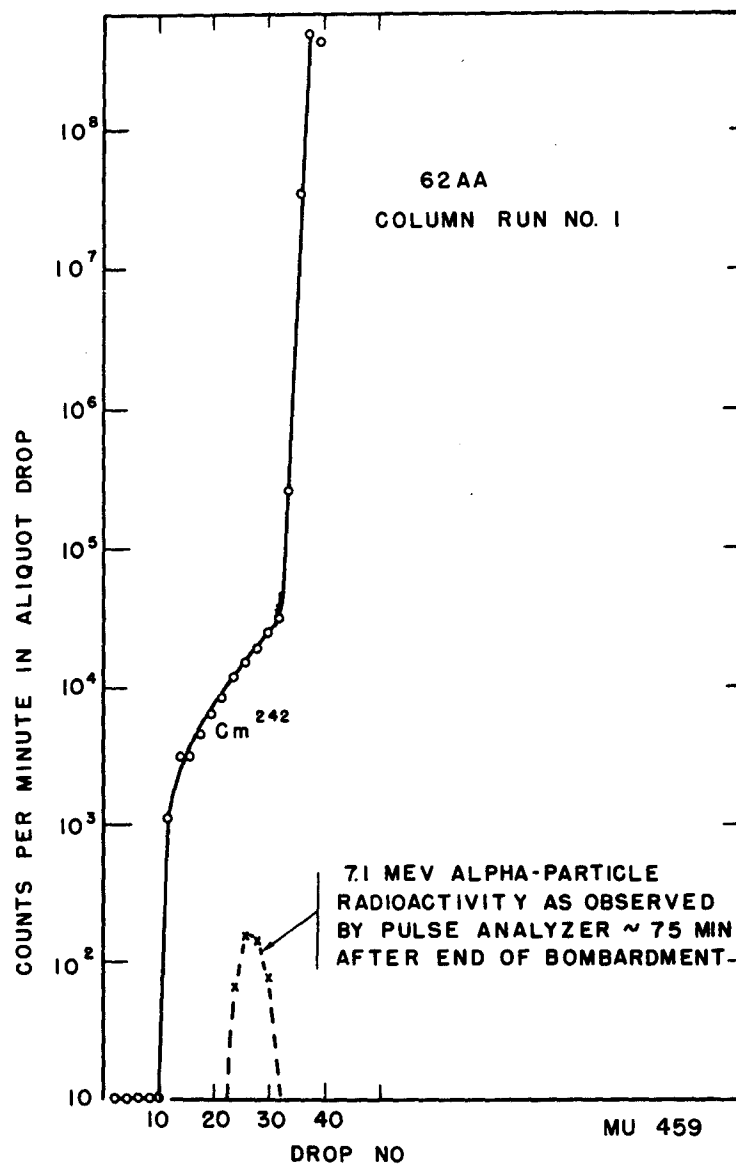


Fig. 3. Results of separation in first Dowex-50 resin column showing small amount of new 7.1-Mev alpha-radioactivity separated ahead of large amount of Cm²⁴² alpha-radioactivity.

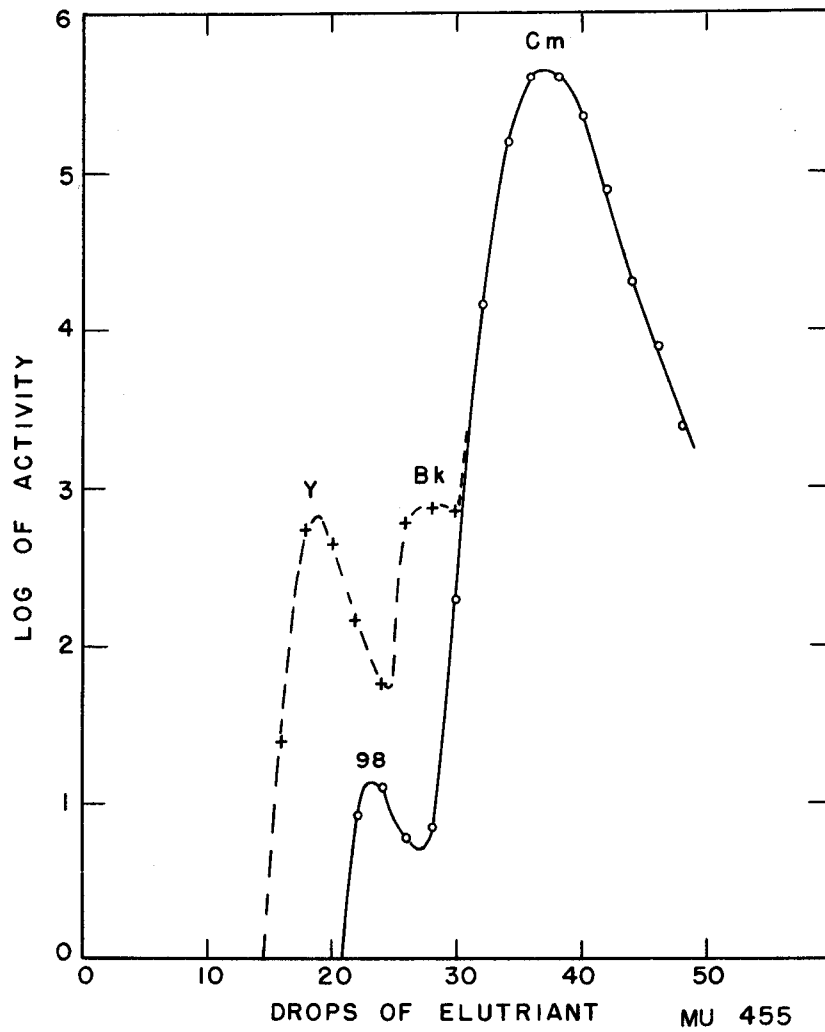


Fig. 4. Results of separation in second Dowex-50 resin column showing 7.1-Mev, 45-minute radioactivity in eka-dysprosium position as compared with isotopes Bk^{243} and Cm^{242} .

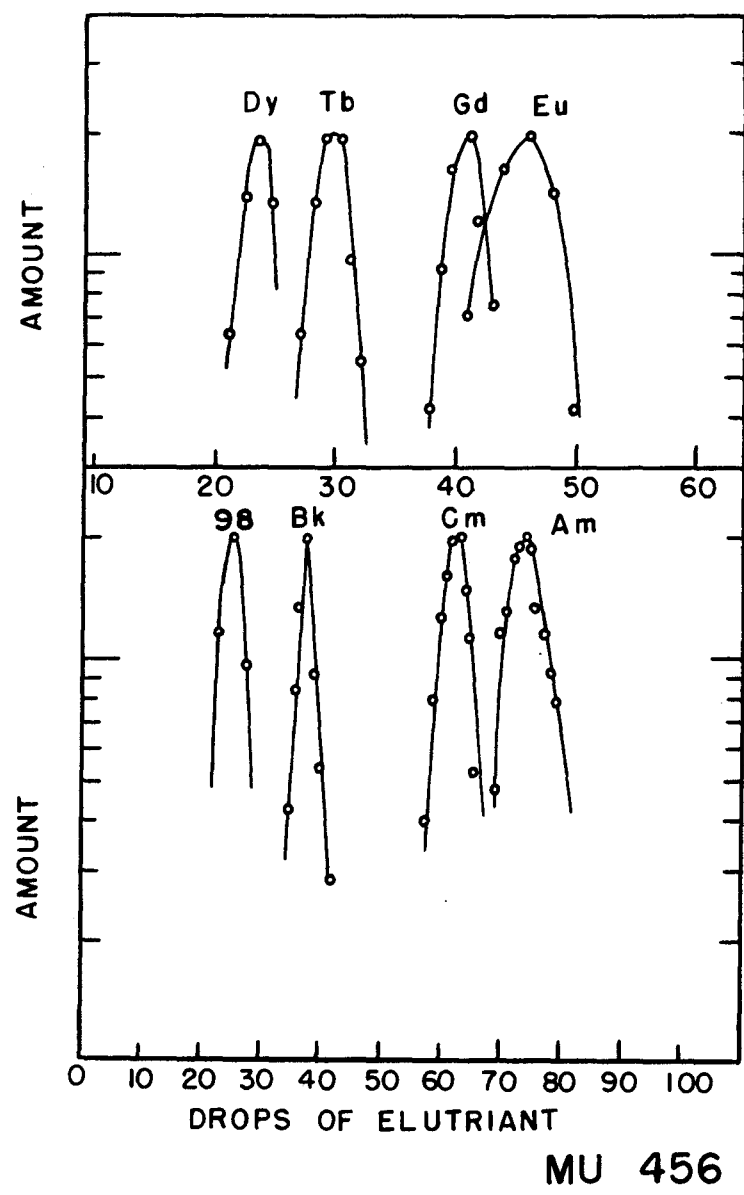


Fig. 5. Summary of relative spacings between actinides and homologous rare earth elements in high temperature Dowex-50 resin columns. The remarkable similarity between homologous actinides and rare earths may be seen.

separated in the fractions containing curium and americium. These fractions were free of rare earth fission products, which were removed from the column much more slowly because of the lesser degree of rare earth complexing with hydrochloric acid.

Additional Chemical Experiments

In some of the later experiments small amounts (approximately 50 counts per minute) of the new alpha-emitter were isolated and further chemical experiments performed in order to characterize somewhat more closely its chemical properties. In these experiments the activity was first separated by carrying on LaF_3 and $\text{La}(\text{OH})_3$ and by the use of ion exchange columns as described above. Because of the small amount of activity produced and the short half-life only a few chemical experiments could be done after each bombardment. The results of these tracer experiments are given below. In all cases the distribution of the 45-minute activity into the various fractions was followed by pulse analysis of the 7.1-Mev alpha-particles.

Several attempts were made to oxidize the 45-minute activity to a higher oxidation state. Carrying with zirconium phosphate was used as a test for oxidation to the (IV) state and lack of carrying on LaF_3 as a test for the (VI) state on the basis of analogy with the behavior of other actinide elements in tracer experiments.

Experiments following oxidation with 0.2M ammonium persulfate for 10 min. at 70°C in a solution 1M in HNO_3 and 0.2M in sulfate showed less than 10 percent carrying with zirconium phosphate and greater than 80 percent carrying with LaF_3 precipitated by adding La^{+++} and making the supernatant 1M in HF.

Experiments following oxidation with sodium bismuthate for 5 min. at 60°C in 5M HNO_3 also showed less than 10 percent carrying with zirconium phosphate and greater than 80 percent carrying with LaF_3 from the supernatant made 3M in HCl.

Although the uncertainties in the tracer experiments are largely due to the small amount of element 98 available, it can be concluded that the oxidation of the 45-minute isotope to the (IV) or (VI) states in aqueous solution even with these strong oxidizing agents is not possible or that the oxidation is slow. In connection with the rapid chemical separations used in this type of work, it is indicated that the introduction of steps to prevent oxidation during the separations are unnecessary.

The chemical properties of element 98 are discussed in somewhat greater detail in another publication.¹⁵

DISCUSSION OF RESULTS

The experiments and results which have been presented provide the evidence for the discovery of an isotope of element 98. That the 45-minute activity observed in the experiments is, beyond reasonable doubt, an isotope of element 98 has been proved by the following essentially independent means:

(1) It separated in the eka-dysprosium position expected for element 98 in elutriant fractions from the Dowex-50 resin columns in which ammonium citrate was used as the eluting agent. The relative spacing between the new activity and its actinide neighbors is remarkably similar to the relative spacings between the homologous rare earth elements.

(2) Its distinctive high alpha-particle energy proves that it must be an isotope of an element above lead in the periodic table. The chemical experiments done on the new isotope distinguish it from all such previously known elements of atomic number 82-97 inclusive. Therefore, the new activity must be an isotope of element 98.

The mass assignment of the 45-minute isotope of element 98 must remain tentative for the present, pending experimental proof. Its assignment as 98^{244} is suggested as being most likely on the basis of estimates using the systematics of

¹⁵ Street, Jr., Thompson, and Seaborg, J. Am. Chem. Soc. (to be published).

alpha-radioactivity as described in the introduction to this paper. The bombardment of Cm^{242} with 33-Mev helium ions is considered to produce directly only the following possible isotopes of element 98 in significant yield--namely, 98^{243} , 98^{244} , and 98^{245} . Of these, only 98^{244} is expected to have a half-life longer than one-half hour unless the electron capture decay is unusually highly forbidden. The observed alpha-particle energy and half-life fit the alpha-decay systematics³ very well for an even-even type nucleus (unforbidden decay) of the element with atomic number 98.

The cross section for the production of the new isotope is about $3 \times 10^{-27} \text{ cm}^2$, based on the observation of the alpha-particles alone. This value is typical of those obtained in the production of other transuranium elements by bombardment with helium ions of energy 30 to 40 Mev. This cross section is sufficiently large to indicate in itself that alpha-particle emission is the principle mode of decay. For example, an electron-capture-to-alpha-decay branching ratio of 10 would require a cross section higher by a factor of 10 than the above and thus a much higher reaction cross section for the (α, xn) reactions than has been observed previously for the production of transplutonium isotopes. It is believed, however, based on estimates using the systematics of radioactivity, that the isotope 98^{244} is unstable toward electron capture decay. Estimates based on the electron capture disintegration energy as determined from closed decay cycles involving estimated energies and on the very rough empirical relationship between half-life and disintegration energy¹² indicate that the half-life of 98^{244} for decay by electron capture should be somewhat longer than that for decay by alpha-emission and hence that the branching ratio toward electron capture decay is less than one.

It is interesting to note that the amount of element 98 which has been isolated has never exceeded a few thousand atoms in any of the bombardments to date.

SUMMARY

Definite identification has been made of an isotope of the element with atomic number 98 through the irradiation of Cm^{242} with approximately 35-Mev helium ions in the Berkeley Crocker Laboratory 60-inch cyclotron. The isotope which has been identified has an observed half-life of about 45 minutes and probably has the mass number 244. The observed mode of decay of the 98^{244} is through the emission of alpha-particles, with energy of about 7.1 Mev, which agrees with predictions. Other considerations involving the systematics of radioactivity in this region indicate that it should also be unstable toward decay by electron capture.

The chemical separation and identification of the new element was accomplished through the use of ion exchange adsorption methods employing the resin Dowex-50. The element 98 isotope appears in the eka-dysprosium position on elution curves containing 4.6-hour Bk^{243} and the bombarded Cm^{242} as reference points--that is, it precedes berkelium and curium off the column just as dysprosium precedes terbium and gadolinium. The experiments so far have revealed only the tripositive oxidation state of eka-dysprosium character and suggest either that higher oxidation states are not stable in aqueous solutions or that the rates of oxidation are slow.

The successful identification of so small an amount of an isotope of element 98 was possible only through having made apparently accurate predictions of its chemical and radioactive properties.

NAME

It is suggested that element 98 be given the name californium (symbol Cf) after the university and state where the work was done. This name, chosen for the reason given, does not reflect the observed chemical homology of element 98 to dysprosium (No. 66) as the names americium (No. 95), curium (No. 96), and berkelium (No. 97) signify that these elements are the chemical homologs of europium (No. 63), gadolinium (No. 64), and terbium (No. 65), respectively.

ACKNOWLEDGMENTS

It is a pleasure to acknowledge the special help in the bombardment of the curium samples of Professor J. G. Hamilton, G. B. Rossi, T. M. Putnam, Jr., M. T. Webb, and the operating crew of the 60-inch cyclotron in the Crocker Laboratory. The successful handling in a safe manner of the radioactivity involved was made possible through the use of the excellent protective equipment provided by Nelson Garden, the members of his Health Chemistry Group, and C. M. Gordon. We wish also to thank E. K. Hulet and G. H. Higgins for their assistance in the experiments. We are happy to acknowledge the continued support and interest of Professor E. O. Lawrence in this work.

This work was performed under the auspices of the AEC.