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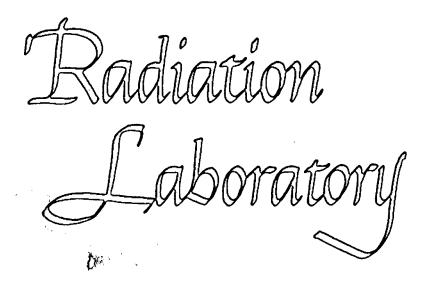
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UNIVERSITY OF CALIFORNIA

Radiation Laboratory Berkeley, California

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BERKELIUM AND CALIFORNIUM

B. B. Cunningham

October 1958

BERKELIUM AND CALIFORNIUM

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Radiation Laboratory and Department of Chemistry University of California, Berkeley, California

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I. Discovery of The Elements

Berkelium and californium, elements 97 and 98, were discovered some eight years ago at the Radiation Laboratory of the University of California — the former by S. G. Thompson, A. Ghiorso and G. T. Seaborg¹ and the latter by Thompson, K. Street, Jr., Ghiorso and Seaborg.²

Neither element now exists in detectable amount as a natural component of the terrestrial environment, although both may have been of significant abundance some four billion years ago, when the earth was formed. However, none of this primordial stock — if, indeed it ever existed — could have survived the long span of time which has since elapsed. The most stable isotope of either element has a half-life of 7 x 10^3 years. The fraction of any original amount of this isotope remaining after 4×10^9 years would be only one part in $\frac{1}{10^{1.6 \times 10^5}}$, a fraction which is much less than one atom of a mass of this isotope equal to the mass of the earth. It would be fruitless, therefore, to search for primordial berkelium or californium in nature.

A short half-life does not necessarily preclude the possibility of the natural existence of a radio isotope, however.

The plutonium isotope of mass 239 has a half-life of only 24000 years—and yet it has been recovered in identifiable (micrograms) amounts from uranium ores. Plutonium-239 exists in such sources because it is being formed continuously by the capture of neutrons by U^{238} and subsequent rapid β^- decays to yield successively Np^{239} and then Pu^{239} . In this process there is a minuscule steady state concentration of the 2.3 day β^- emitting Np^{239} .

The natural neutron flux in uranium ores is exceedingly small, however, and isotopes which require several successive neutron captures for their formation (as is true of the isotopes of Bk and Cf) cannot be sustained at identifiable levels in these deposits. For such elements, therefore, synthesis is a necessary prelude to discovery.

This work was performed under the auspices of the U.S. Atomic Energy Commission.

The two principal methods used for the synthesis of the transuranium elements are neutron irradiation and charged particle bombardment. In most cases the latter method has been used in the initial preparation and identification of a new transuranium element, in spite of the fact that only relatively small numbers of atoms can be produced in this way. This disadvantage is more than compensated by the relatively short half-lives and hence high specific radioactivities of the isotopes produced, by the low total radioactivity of the target which is favorable to rapid chemical processing, and by the fact that the bombardments may be completed within a few hours.

The initial syntheses of berkelium and californium were effected by charged particle bombardment, the projectiles being helium ions of 35 million electron volt energy, produced by the Berkeley 60-inch cyclotron, and the target materials ${\rm Am}^{241}$, in the case of berkelium, and ${\rm Cm}^{242}$, in the case of californium.

The new elements were formed by the nuclear reactions:

$$95^{\text{Am}^{241}} + 2^{\text{He}^{4}} \longrightarrow 97^{\text{Bk}^{243}} + 20^{\text{n}^{1}}$$
 (1)

$$_{96}^{\text{Cm}^{242}} + _{2}^{\text{He}^4} \longrightarrow _{98}^{\text{Cf}^{245}} + _{0}^{\text{n}^1}$$
 (2)

where on stands for a neutron, zero atomic number and mass 1. Other reactions, such as fission of the target nuclei also occurred in the bombardments, but need not concern us here.

It was possible to predict in advance of the actual bombardments some very rough figures for the yields of the desired isotopes. Thus, the probability of reaction (1) occurring between one incident helium ion and one atom of Am²⁴¹ in the target was estimated to be about 10⁻²⁶. In the bombardment the probability would be increased in proportion to the number of Am²⁴¹ atoms in the target and the number of helium ions furnished by the beam — the latter being proportional to the beam current and the duration of bombardment. However, it is useless to extend the bombardment for a time much greater than 1.5 times the half-life of the desired product isotope, for by then a steady state is reached in which the rate of production of the isotope is balanced by

its rate of decay. The rate of decay that is the number of atoms decaying per unit time of N atoms of an isotope of half-life, T 1/2, is equal to N $\frac{0.693}{T \cdot 1/2}$.

If R is the rate of production (atoms formed per unit time) then at steady state: $N = \frac{R \times T}{0.693}$.

R is readily calculated from the reaction probability, the number of target nuclei and the number of projectile nuclei which impinge on the target per unit time.

In the bombardment of Am^{241} , approximately 10 mg of the isotope were used for the target, which was exposed to a helium ion current of about 2 microamperes. Therefore

 $R = 10^{-26} \times \frac{1 \times 10^{-2}}{241} \times 6 \times 10^{23}$ (atoms of Am in target)

x 6 x 10^{12} (He⁺⁺ ions per second) \cong 1.5 x 10^6 (atoms Bk²⁴³ formed per second) At steady state, the number of atoms N of Bk²⁴³ in the target would be:

$$N = \frac{1.5 \times 10^6 \times T \, 1/2}{0.693}$$

On the basis of an expected half-life of Bk^{243} of around an hour, or 3600 seconds the expected steady state number of Bk^{243} atoms was calculated to be about 7 x 10.

In actuality the half-life of Bk^{243} proved to be about $4\ 1/2$ hours, so that about $3\ x\ 10^{10}$ atoms were synthesized in the initial bombardment. The isotope decays mainly by the capture of an orbital electron — a process which is detected with only about 2% efficiency. From the amount of berkelium produced about $6\ x\ 10^8$ decay events were thus available for tracing the course of the element during its chemical separation and identification.

Because of a smaller reaction probability and a smaller amount of target material, only about 7×10^5 atoms of Cf²⁴⁵ were produced in the initial synthesis of this element.

For both berkelium and californium the technique of identification rested on the postwar development of the extremely powerful, yet simple technique of ion exchange separation. I is fair to say that without the developments of this technique the discoveries of the heavier transuranium elements would have been immensely more difficult, and undoubtedly greatly delayed.

The ion exchange technique is now so familiar that it need not be described here.

It will suffice to show, as is done in Fig. 1, below, the elution sequence of a number of lanthanide and actinide elements from the cation exchange resin, Dowex-50, using citrate ion as eluting agent.

The important feature of these curves is that within each series the tripositive ions of both lanthanides and actinides exhibit an elution order which is the reverse of the order of atomic number. It will also be noted that the spacings between the successive peaks follow a well defined pattern. The spacings, in the case of both series, may be correlated with the variation of ionic radius with atomic number.

Since, in heavy element bombardments, the lanthanide elements are formed as fission products, it is necessary to effect a group separation of the lanthanides from the actinides—otherwise the elution positions of the two series would overlap. This separation can be accomplished by ion exchange, using concentrated solutions of chloride ion as eluant, owing to the greater stability of the polychloride complexes of the actinides as compared with the lanthanides.

Chemical processing following a bombardment includes a group separation of the two series, followed by separation of the individual actinide elements.

A reproduction of the original elution curve of the actinide fraction recovered from the first helium ion bombardment of ${\rm Am}^{241}$ is shown. Delical

Berkelium appears in the expected position, just ahead of curium and americium.

In the discovery of californium, berkelium was added as tracer before the elution, the californium peak then appearing just ahead of the berkelium.

Since the initial discovery of these two elements, a number of isotopes other than ${\rm Bk}^{243}$ and ${\rm Cf}^{245}$ have been prepared by various techniques. These are listed in Table 1 below.

Table 1

		Isotopes of berkelium and d	iolifornium
1		Isotopes of berkeli	
Isotope	Half-life	Mode of disintegration a	Source
Bk ²⁴³	4.5 h	E.C. (> 99%) α (0.15%)(6.72,6.55,6.20)	$Am^{241}(\alpha,2n)$
Bk ²⁴⁴	4.4 h	E.C. (> 99%) α (6 x 10 ⁻³ %)(6.66)	$Am^{241}(\alpha,n)$
Bk ²⁴⁵	4.95 d	E.C. (> 99%) α (0.1%)(6.33,6.17,5.89)	$Am^{243}(\alpha,2n)$
Bk ²⁴⁶	1.9 d	E.C.	$Am_{244}^{243}(\alpha,n)$ Cm (α,pn)
Bk ²⁴⁷	7 х 10 ³ у	α (5.67,5.50,5.30)	Cf_{244}^{247} E.C. decay $Cm_{244}^{244}(\alpha,p)$
Bk ²⁴⁸	16 h	β ⁻ (70%)(0.65) E.C. (30%)	$\frac{2^{47}(n, \gamma)}{2^{46}(\alpha, p)}$
Bk ²⁴⁹	2 290 Cd	β ⁻ (> 99%)(0.09) α (10 ⁻³ %)(5.40,5.08)	Pu ²³⁹ multiple neutron capture U ²³⁸ multiple neutron capture
Bk ²⁵⁰	3.13 h	β¯ (1.9,0.9)	Bk ²⁴⁹ (n,γ)
		Isotopes of californi	um
Cf ²⁴⁴	25 m	α (7.17)	Cm ²⁴² (α,2n) U ²³⁸ (C ¹² ,6n) Pu ²³⁹ (Be ⁹ ,4n)
Cf ²⁴⁵	- 1,4. m	E.C. (66%) α (34%)(7.11)	$Cm^{242}(\alpha,n)$ $Cm^{244}(\alpha,3n)$ $U^{238}(C^{12},5n)$ $Pu^{239}(Be^{9},3n)$
Cf ²⁴⁶	35.7 h	α (6.753,6.711) β stable	$C_{m}^{244}(\alpha, 2n)$ $U^{238}(C_{12}^{12}, 4n)$ $U^{238}(N^{14}, p5n)$ $P_{12}^{239}(R_{e}^{9}, 2n)$
Cf ²⁴⁷	2.4 h	E.C.	$Cm^{244}(\alpha,n)$ $U^{238}(N^{14},p4n)$
Cf ²⁴⁸	250 d	α (6.26) β stable	$U^{236}(N^{14},p3n)$ $Cm^{245}(\alpha,n)$
Cf ²⁴⁹	470 у	α (6.190,5.91,5.82) β stable	Bk ²⁴⁹ β-decay
Cf ²⁵⁰	9.3 у	α (6.024,5.980) β stable	Pu ²³⁹ multiple neutron capture Bk ²⁴⁹ (n, γ) Bk ²⁵⁰ β

Table 1 (cont'd.)

		Isotopes of berkelium and c	alifornium	
Isotopes of californium				
Isotope	Half-life	Mode of disintegration a	Source	
Cf ²⁵¹	660 у	α	Pu ²³⁹ multiple neutron capture	
Cf ²⁵²	2.2 y α 66 y	α (6.112,6.069) α/S.F. ~30	Pu ²³⁹ multiple neutron capture U ²³⁸ multiple neutron capture	
	spontaneous fission	β stable		
Cf ²⁵³	20 d	β	Pu ²³⁹ multiple neutron capture	
Cf ²⁵ 4	55 d	spontaneous fission β stable	E ²⁵⁴ E.C. decay Pu ²³⁹ multiple neutron capture	
a _{En on our}	of modiation	in million electron volta.		

Energy of radiation in million electron volts; E.C. = electron capture; S.F. = spontaneous fission.

II. Chemical Properties of Bk and Cf Deduced from Tracer Studies.

A few atoms of an element may suffice for a limited investigation of its chemical properties if it is available as a radioisotope. The energy released in a single decay event usually may be readily detected and characterized. Thus the presence of only one such atom in a sample under investigation can be revealed by radioassay. In this case, of course, the probability of decay must be such that it can be expected to occur in a reasonable period of observation. On the other hand, it must not be so large that decay is likely before the preparation of the sample is completed.

Chemical investigations carried out by such methods are called "tracer" studies. They can serve to reveal directly only one property of the element under investigation—its relative preference for one kind of environment as compared with another—that is its phase distribution.

Nevertheless a great deal may be learned by inference from such studies, including the identity of various oxidation states, approximate values of oxidation-reduction potentials of couples involving different oxidation states, the composition and stability of complex ions, and relative volatility as the element and in the form of various compounds.

The initial chemical identifications of berkelium and californium demonstrated the stability of their tripositive oxidation states in aqueous solution, since the elements appeared in their proper order in the elution sequence with other tripositive actinide ions.

In later experiments with tracer amounts of berkelium, it was shown that the tripositive ion could be extracted into benzene as a complex with TTA (thenoyl trifluoro acetone). The extraction coefficient was found to be some 15 times greater than that of curium.

Californium in the elemental or metallic state was observed to be much more volatile than uranium, since it could be readily volatilized from the latter metal at about 1200° C.

The electronic configurations of Bk⁺³ and Cf⁺³, predicted on the basis of Seaborg's actinide hypothesis, suggested the desirability of early experiments on the oxidation-reduction behavior of these ions. Thus the predicted electronic configurations for Bk⁺³ and Cf⁺³ were 5f⁸ and 5f⁹ respectively (beyond the radon core.) Loss of a single electron by Bk⁺³ and of two electrons by Cf⁺³ would

yield the especially stable half-filled subshell configuration of 5f7.

Attempts to prepare higher oxidation states corresponding to these configurations were begun in 1950.

In one such set of experiments tracer amounts of Bk^{+3} were added to acid solutions containing a few milligrams of Zr^{+4} .

The solutions were then treated with various oxidizing agents such as Br_2 , Cl_2 , BrO_3^- and $\mathrm{Cr}_2\mathrm{O}_7^-$. After about a half hour the zirconium was precipitated as $\mathrm{Zr}_3(\mathrm{PO}_4)_4$ by the addition of phosphoric acid. The precipitate and supernatant solution were separated and each fraction examined for berkelium by radiometric assay. With Br_2 and Cl_2 only a few percent of the berkelium was found in the precipitate whereas with BrO_3^- and $\mathrm{Cr}_2\mathrm{O}_7^-$ most of the berkelium was found in this fraction. The zirconium phosphate precipitate is known to coprecipitate +4 ions selectively from a mixture of +3 and +4 ions and hence it could be concluded that oxidation of berkelium probably to the +4 testate - had occurred in the presence of BrO_3^- and $\mathrm{Cr}_2\mathrm{O}_7^-$ but not in the presence of Br_2 or Cl_2 .

In later experiments, using zirconium phenylarsonate as carrier (selective for the coprecipitation of +4 ions in a mixture of actinide ions in the III, TV, V, and VI oxidation states) the dxidation state of the oxidized berkelium was definitely identified as +4. Moreover by studying the percent of oxidation of berkelium as a function of the ratio of Ce^{+4} to Ce^{+3} concentration in solution it was possible to fix the potential of the $Bk^{+3} = Bk^{+4} + e^-$ couple as -1.62 \pm 0.06 v.

However, in similar studies of the oxidation-reduction behaviour of californium no evidence of oxidation beyond the +3 state has ever been found, not even by such a powerful oxidizing agent as peroxydisulfate. Probably the higher oxidation states of californium are themselves such powerful oxidizing agents that they are instantly reduced by water.

III. <u>Isolation of Macroscopic Amounts of Berkelium and Californium and</u> Observation of Macroscopic Properties.

A complete study of the chemistry of an element is not possible by tracer methods alone, since for the determination of properties such as melting point, absorption spectrum, heat capacity, etc. large aggregates of atoms or molecules are required.

Although by a suitable refinement of experimental techniques it is possible to determine such macroscopic properties with quantities of material very much smaller than those used conventionally, at the present time these minimal amounts are still of the order of a microgram.

It is not feasible, in general, to prepare such amounts of the transuranium elements by charged-particle bombaraments — at least with existing cyclotron beam currents. Moreover the short half-lives of the isotopes so prepared would render them thermally unstable as macroscopic aggregates.

Longer lived isotopes may be made, however, by multiple neutron capture on lighter target materials. Such multiple neutron capture leads eventually to a mass-to-nuclear charge ratio which is favorable for β emission. This emission gives a daughter nucleus which is one atomic number higher than the parent--i.e., to the next higher element. Further neutron capture by the daughter again lends to β decay, and so on. Such a chain, leading to the production of isotopes of Bk and Cf is outlined below:

Pu²³⁹
$$(n,\gamma)$$
 Pu²⁴⁰ (n,γ) Pu²⁴¹ (n,γ) Pu²⁴² (n,γ) Pu²⁴³

Pu²⁴³ $\frac{\beta}{5.0 \text{ hr}}$ Am²⁴³ (n,γ) Am²⁴⁴ $\frac{\beta}{26 \text{ m}}$ Cm²⁴⁴ (n,γ) Cm²⁴⁵
 (n,γ) Cm²⁴⁶ (n,γ) Cm²⁴⁷ (n,γ) Cm²⁴⁸ (n,γ) Cm²⁴⁹

Cm²⁴⁹ $\frac{\beta}{65 \text{ m}}$ Bk²⁴⁹ (n,γ) Bk²⁵⁰ $\frac{\beta}{3.13 \text{ hr}}$ Cf²⁵⁰ (n,γ) Cf²⁵¹
 (n,γ) Cf²⁵² (n,γ) Cf²⁵³ (n,γ) Cf²⁵⁴.

The expression (n,γ) indicates neutron capture followed by gamma emission (which does not effect the nuclear charge).

In order to obtain appreciable yields of the heavier isotopes in a reasonable period of time a very high neutron flux is required, since the amount of a heavy isotope produced per unit time has an exponential dependence on the flux, the exponent being equal to the difference in mass between the target nucleus and that of the desired isotope. In these irradiations, yields are further reduced by the competing reaction of fission, which occurs extensively in neutron irradiations of heavy element isotopes. In spite of these difficulties, macro amounts of the very heavy isotopes of the transuranium elements are produced in this way.

As an example, a neutron irradiation for the purpose of providing macro amounts of berkelium and californium was initiated in 1953, the target material being 10 grams of Pu²³⁹. This was placed in a position in the Materials Testing Reactor at Arco, Idaho, where the neutron flux amounted to about 10¹⁴ neutrons per cm² per second. The irradiation was continued for a period of 5 years, during which time approximately 90% of the heavy element isotopes had undergone fission.

Among the heavy isotopes remaining, however, there were about 0.6 μg of berkelium and 1.2 μg of californium. Chemical processing eventually yielded these elements in separated and purified form.

Using these materials, the author, in collaboration with Dr. S. G. Thompson, Llad Phillips and Ray Gatti initiated studies of the macroscopic properties of these elements, in the early part of this year.

We decided to attempt three kinds of observations on each element: (1) The determination of the absorption spectra of the aqueous tripositive ions, (2) Measurements of the magnetic susceptibilities of the ions, and (3) An x-ray diffraction study of the oxides. We were interested in the first two kinds of observations because of the information to be obtained concerning the electronic configurations of the ions, and in the last because of the possibility of observing higher oxidation states in solid compounds. To date we have carried out experiments relating to only the first two of these objectives.

In developing the experimental methods to be utilized for this research we were particularly concerned that our techniques should in no case involve any real risk of loss of a major portion of either of the samples.

For example, in planning for the observation of the absorption spectrum, the method finally chosen was one which did not require any extra manipulation

of the samples. This was done by attaching to the lower end of the ion exchange column which was to be used in any event for the final purification of each element, a capillary absorption cell, thru which the purified sample passed on its way to its final container. Light transmitted thru the absorption cell was examined with a bench spectrometer. This arrangement is shown in diagrammatic form in Fig. 3.

The equipment was tested in preliminary experiments in which small quantities of americium and neodymium were eluted by the same technique as that utilized subsequently for berkelium and californium.

We had no difficulty in observing the absorption spectra of the ions of these elements as they were eluted from the resin and passed thru the absorption cell.

From these preliminary experiments we were able to conclude that any absorption lines of Bk^{+3} or Cf^{+3} , of molar extinction coefficient \geq 20 within the range from 4500 to 7500 A should be detectable with our equipment.

In careful observations on both Bk^{+3} and Cf^{+3} no absorption in the indicated range was detected. In later experiments with Cf^{+3} , done in collaboration with J. G. Conway, the spectral range was extended by using a grating spectrograph and photographic detection. Two broad, weak absorptions were observed, centering at 7800 and 8300 A respectively. It is tentatively suggested that these are due to transitions from the $^{6}\mathrm{H}_{15/2}$ ground state to the $^{6}\mathrm{P}_{5/2}$ and $^{6}\mathrm{P}_{3/2}$ upper states analogous to the transitions at slightly shorter wave lengths observed in Dy^{+3} , the rare earth analog of Cf^{+3} .

It must be borne in mind however, that if the transition probabilities between energy levels in the 5f subshell have dropped at berkelium and californium to values comparable to those observed with the rare earths, we would not have been able to observe such weak absorptions even if they occurred in the visible spectrum.

For the measurement of the magnetic susceptibility of Bk^{+3} and Cf^{+3} it was again necessary to develop special techniques. In order to provide a desirable diamagnetic diluent for the paramagnetic heavy element ions and at the same time to afford a method for the mechanical manipulation of the samples, it was decided that the measurements should be made with the Bk^{+3} or Cf^{+3} adsorbed on a single small bead of cation exchange resin, which could then be examined in a

magnetic field. Of course, it would be necessary in preliminary experiments to determine the appropriate corrections for the magnetic behaviour of the head alone.

The magnetic susceptibility of a solid sample usually is determined by some modification of the Faraday method, in which the sample is exposed to the action of an inhomogeneous magnetic field.

The force experienced by the sample under these circumstances is given by the relation

$$\mathbf{F} = \mathbf{m} \times_{\mathbf{g}} \mathbf{H} \frac{\mathbf{dH}}{\mathbf{dx}}$$
,

where m is the mass of the sample, Xg its gram susceptibility,H is the strength of the magnetic field and $\frac{dH}{dx}$ the field gradient. F is in the direction of the field inhomogeneity.

Since H $\frac{dH}{dx}$ is difficult to measure accurately it is customary to calibrate the apparatus with a known mass of material of known susceptibility. The measurement of the susceptibility of a sample under investigation is then reduced to a measurement of the relative forces experienced by known weights of the standard and sample.

For very small samples of material, such as we were planning to use, these forces would naturally be very small. We could calculate in advance that they would be of an order of (5 to 20) \times 10⁻⁵ dynes. Precision to 1% required the capability of detecting a force of 5 \times 10⁻⁷ dynes.

We proposed to achieve the required sensitivity by arranging the magnet pole pieces so that the magnetic force on the bead would be directed in a horizontal plane, and suspending the bead from a long, light and very flexible vertical support. Since fibers of fused silica may be drawn down to micron thickness, this was chosen for the suspension. Practical considerations limited its length to about 30 cm. The mass of the bead was about 50 μg . We could calculate the displacement of the suspended bead under the action of a horizontal force of 5 x 10^{-7} dynes:

displacement in mm =
$$\frac{300 \text{ mm x 5 x 10}^{-7} \text{ dynes}}{5 \text{ x 10}^{-5} \text{ gm x } 980 \frac{\text{dynes}}{\text{gm}}} = 3 \text{ x } 10^3 \text{ mm}.$$

In order to detect this displacement, it was necessary to observe the movement of the bead with a microscope. A microscope of moderate magnification, fitted with a filar micrometer, was found to be satisfactory for the purpose.

The added requirement that the measurements should cover a wide range of temperatures determined the final form of the equipment, which is illustrated in Fig. 4.

The experimental results on Cf⁺³ are given in Fig. 5.

The data indicate that the susceptibility conforms rather well to the Curie law, (susceptibility proportional to the reciprocal of the absolute temperature) but are best fitted by a Curie-Weiss relationship, with a small constant correction \triangle added to the absolute temperature. Similar data were obtained for Bk⁺³. The effective magnetic moments calculated from the susceptibilities at various temperatures are concordant within experimental error (about $\pm 10\%$) and give calculated values of 8.7 and 9.2 Bohr magnetons for Bk⁺³ and Cf⁺³ respectively.

These moments are plotted in Fig. 6, along with the theoretical values derived from the three different assumptions: (a) the elements belong to a "d" transition series, the ions exhibiting "spin only" magnetism in condensed phases, (b) the elements belong to an "f" transition series, there being complete quenching of the orbital momentum, and (c) the elements are members of an "f" transition series, there being no quenching of the orbital momentum in condensed phases.

Shown on this same plot are experimental values of this magnetic moments of lighter actinide ions.

The experimental values clearly conform best to the assumptions of (c) above, although there is noticibly more deviation from the theoretical moments than in the case of the rare earths, where the experimental values conform to (c) within a few percent.

The magnetic data clearly indicate, however, that the electronic configurations of Bk^{+3} and Cf^{+3} very probably are $5f^{8}$ and $5f^{9}$ respectively.

In the above description of the work with berkelium and californium, no particular mention was made of the isotopic compositions of the samples. Nevertheless, isotopic composition may have an important bearing on the selection of techniques used for chemical studies of macroscopic properties.

The berkelium sample was composed mainly of Bk^{249} , which decays almost entirely by the emission of 0.1 Mev β particles with a half life of about 300 days. A very slight (\sim 10⁻³%) decay by alpha particle emission occurs simultaneous, as does decay by spontaneous fission, with the release of neutrons. The latter process is of negligible magnitude, however.

The sample of californium, on the other hand contained a large proportion $(\sim65\%)$ of Cf^{252} , with an alpha half-life of about 2.3 years and a half life for spontaneous fission of only 66 years. The neutron flux from the spontaneous fission is a serious potential hazard to the experimentor. With the very small amount of Cf^{252} in our sample, the neutron flux did not prove to be a serious disadvantage. In fact, special counters were developed to count these neutrons, as an aid in radioassay measurements. In any future work with more than a few micrograms of this isotope, it will be necessary to do all manipulations by remote control — a requirement difficult to meet in working on a submilligram scale.

The other isotopes present in the sample $- \text{Cf}^{249}$, Cf^{250} , Cf^{251} — did not contribute substantially to the total spontaneous fission activity.

 ${\rm Cf}^{249}$, the daughter of Bk 249 is much more suitable for chemical studies, since it has a 360 year half life for alpha emission and a 400 year half life for spontaneous fission.

In conclusion we may note that einsteinium probably marks the end of the road so far as the possibility of isolation of any higher elements in macro amounts is concerned. No isotope of any element beyond einsteinium has sufficient nuclear stability for separation in macroscopic quantity.

However, further development and refinement of experimental techniques may be expected to permit continuing exploration of the macroscopic properties of the transuranium elements through einsteinium, while studies of higher elements will be limited to those properties which may be deduced by tracer techniques.

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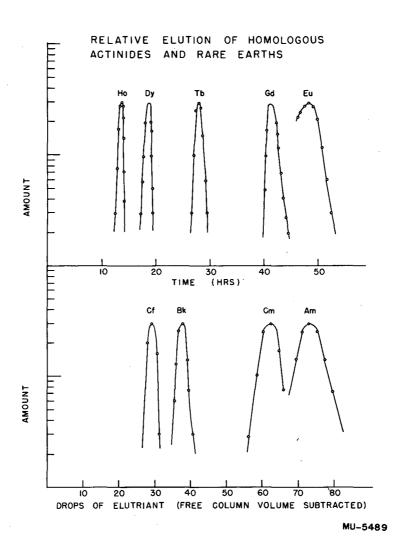


Fig. 1. Elution of lanthanides and actinides from Dowex-50, with citrate solution.

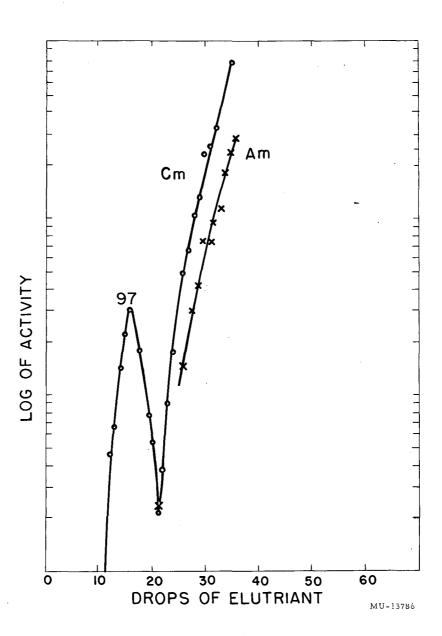
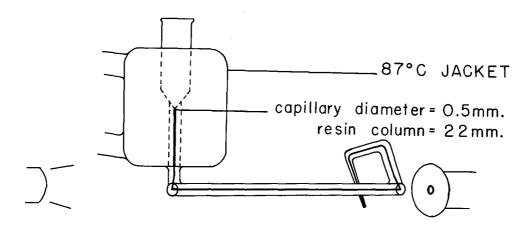


Fig. 2. Reproduction of original elution curve illustrating the discovery of element 97 (berkelium).

COMBINED ION EXCHANGE COLUMN AND ABSORPTION CELL



LAMP ABSORPTION CELL: SPECTROMETER length = 100 mm. capillary diameter = 0.3 mm.

MU-1493

Fig. 3. Capillary absorption cell.

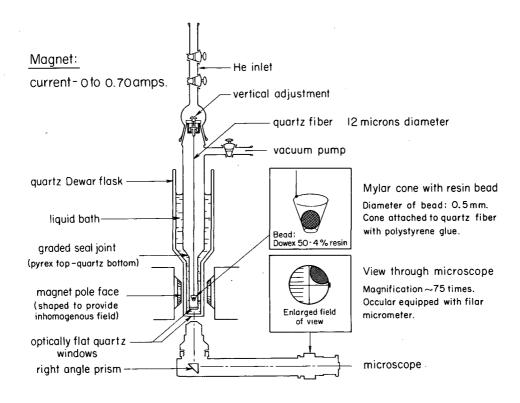


Fig. 4. Magnetic susceptibility apparatus.

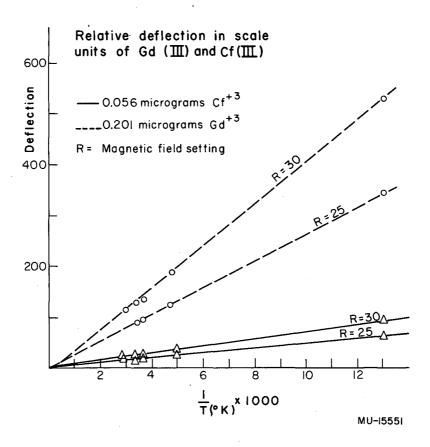


Fig. 5. Relative susceptibilities of Gd^{+3} and Cf^{+3} as a function of temperature.

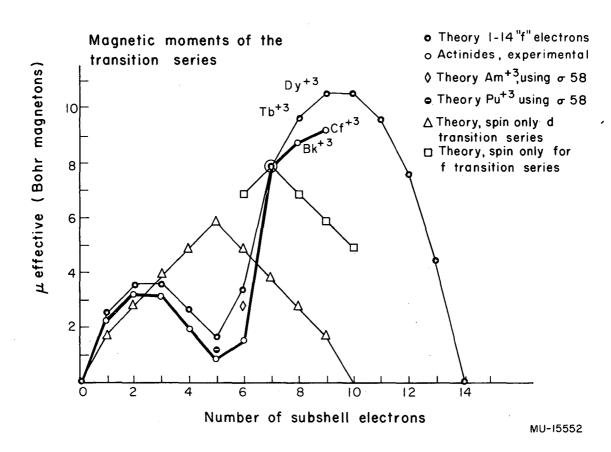


Fig. 6. Effective magnetic moments of various actinide ions, compared with theoretical values.