**RADIOCHEMICAL STUDIES OF NEUTRON DEFICIENT
ACTINIDE ISOTOPES**

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DEDICATION

This thesis is dedicated to some of the people who have had a profound effect upon my life:

To Shirley for the trips to Echo Lake, her friendship and encouragement, and for proving "it could be done";

To my parents, Lee and Willie, who have continually given me encouragement and support, and who have always taken pride in my accomplishments;

And finally, to Ken, my fiance, for all his confidence in me, his understanding, and his love.

There is no heavier burden than a great potential.

- Linus (1970)

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ABSTRACT

The production of neutron deficient actinide isotopes in heavy ion reactions was studied using alpha, gamma, X-ray, and spontaneous fission detection systems. A new isotope of berkelium, ^{242}Bk , was produced with a cross-section of $\sim 10\mu\text{b}$ in reactions of boron on uranium and nitrogen on thorium. It decays by electron capture with a half-life of 7.0 ± 1.3 minutes. The alpha-branching ratio for this isotope is less than 1% and the spontaneous fission ratio is less than 0.03%.

Studies of (Heavy Ion, pxn) and (Heavy Ion, αxn) transfer reactions in comparison with (Heavy ion, xn) compound nucleus reactions revealed transfer reaction cross-sections equal to or greater than the compound nucleus yields. The data show that in some cases the yield of an isotope produced via a (H.I., pxn) or (H.I., αxn) reaction may be higher than its production via an xn compound nucleus reaction. These results have dire consequences for proponents of the " $Z_1 + Z_2 = Z_{1+2}$ " philosophy. It is no longer acceptable to assume that (H.I., pxn) and (H.I., αxn) product yields are of no consequence when studying compound nucleus reactions.

No evidence for spontaneous fission decay of ^{228}Pu , ^{230}Pu , ^{232}Cm , or ^{238}Cf was observed indicating that strictly empirical extrapolations of spontaneous fission half-life data is inadequate for predictions of half-lives for unknown neutron deficient actinide isotopes.

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1. INTRODUCTION

A. Background

When an energetic charged particle approaches a resting target nucleus, a wide variety of reactions can take place. The vast majority of the projectiles will lose energy by interactions with the atomic electrons of the target material. When a particle passes close enough to the target nucleus to be repelled by Coulomb forces (assuming a positively charged particle) the particle will experience a change in direction and a small loss in kinetic energy (Coulomb Scattering). Projectiles following a grazing trajectory experience the short-range nuclear forces and are either elastically or inelastically scattered. Those that suffer only a small loss in kinetic energy and leave the target nucleus in the ground state are elastically scattered while those leaving the target in an excited state (resulting in a loss of kinetic energy by the projectile) are inelastically scattered. In a few cases the projectile may undergo a nuclear reaction with the target nucleus. That is, all or part of the projectile may be captured by the nucleus; or the projectile may absorb or strip away part of the target nucleus. These compound nucleus (complete fusion) and transfer (incomplete fusion or stripping) reactions are the interactions of primary concern in this work.

A number of phenomena characterize compound nucleus formation.¹⁻³ A primary requirement for the existence of a compound nucleus is that all the nucleons from the projectile lose their previous individual and collective characteristics and take on the new characteristics of

the compound system. The kinetic energy of the system is randomly distributed among all the permissible degrees of freedom; the incident nucleons have "forgotten" their origin. Due to the uniform energy distribution, none of the nucleons has enough energy to escape immediately, giving the compound nucleus a long lifetime ($10^{-14} - 10^{-18}$ seconds) in relation to the time necessary for a nucleon to travel through a nucleus ($10^{-20} - 10^{-23}$ seconds)⁴.

The compound nucleus will be in a state of high excitation energy equal to the binding energy of the absorbed projectile to the nucleus(Q) plus the center of mass kinetic energy of the projectile. The newly formed nucleus will recoil in the direction determined by the incoming particle with an energy $E_R = E_i \frac{A_{proj}}{A_{proj} + A_{tgt}}$ where A = mass, E_i = incident projectile energy.

The high excitation energy leads to a finite lifetime due to the statistical fluctuations of energy distribution which can momentarily concentrate enough energy on a single nucleon or a cluster of nucleons to allow it to escape. In most cases, the excited compound system is capable of emitting a number of particles - n, p, d, α - and/or gamma radiation. Since neutrons do not have to overcome a Coulomb barrier, they are the most easily emitted particles, especially from high Z nuclei. While gamma emission is energetically favorable, it is a slow process compared with particle emission. Thus, as long as the excitation energy remains greater than the binding energy of a particle, particle emission will preferentially occur over gamma emission. Once the excitation energy is below the particle-binding energy, gamma emission will become the primary mode of deexcitation. Due to the equilibration

and statistical nature of the concentration of energy on the individual nucleons, particles are emitted isotropically in the center of mass system with relatively low kinetic energy.

For high Z and A and especially for high angular momentum compound nuclei, fission becomes the dominant deexcitation mode. This is due to the decreasing fission barriers of high A (> 150) nuclei. Estimates for the competition between neutron emission and fission can be made by comparing the crude expressions for fission (Γ_f) and neutron emission (Γ_n) widths taken from Vandenbosch and Huizenga⁵ and Lefort³:

$$\Gamma_n = C_n \exp \left(-\frac{E^* - B_n}{T} \right) \quad \Gamma_f = C_f^* \exp \left(-\frac{E^* - B_f}{T} \right)$$

where C_n and C_f^* are constants, E^* is the excitation energy, B_n is the neutron binding energy, B_f is the fission barrier at the saddle point and T is the nuclear temperature. $\exp \left(-\frac{E^* - B_n}{T} \right)$ is the approximate expression for the average level density of the residual nucleus after emitting a neutron having binding energy B_n ; and $\exp \left(-\frac{E^* - B_f}{T} \right)$ is proportional to the average level density at the saddle point. The resulting ratio is: $\frac{\Gamma_n}{\Gamma_f} = C \exp \left(\frac{B_f - B_n}{T} \right)$ where C is approximately equal to $\frac{2TA^{2/3} 2\pi r^2}{h^2} \approx \frac{2TA^{2/3}}{10} \text{ MeV}^{-1}$ (5). This expression is valid over only a limited range of excitation energy and where B_n and B_f are of comparable

magnitude. For the heavy elements, there is evidence that $\frac{\Gamma_n}{\Gamma_f}$ does not vary dramatically with excitation energy.⁶⁻⁸ Angular momentum in this derivation can be simply accounted for by subtracting the rotational energy ($E_r = \frac{J^2 h^2}{2I}$) from the excitation energy, E^* . The level density with spin J is roughly the same as the level density without spin

restriction but at a lower excitation energy, $E^* = \frac{J_n^2}{2I}$, where I is the moment of inertia. Since I is smaller for a sphere than for an ellipsoid, the rotational energy for neutron emission, E_{R_0} (spherical shape), is larger than that for fission, E_{R_S} (saddle point shape). The neutron width to fission width ratio then becomes:

$$\frac{\Gamma_n}{\Gamma_f} = C \exp \left(\frac{-B_n - E_{R_0} + B_f + E_{R_S}}{T} \right)$$

Finally, a rotating liquid drop fission barrier can be defined as

$B_{f_R} = B_f - (E_{R_0} - E_{R_S})$. Thus,

$$\frac{\Gamma_n}{\Gamma_f} = C \exp \left(\frac{B_{f_R} - B_n}{T} \right) \quad \text{becomes small}$$

when $B_n = B_f$ and negligible when $B_f = 0$. Therefore, high angular momentum, in addition to decreasing the total fusion cross-section,⁹⁻¹¹ leads to high fission yields of those compound nuclei that are formed.

In the very heavy region ($A \gtrsim 200$) the fission barriers without rotational considerations are already below 15 MeV and B_f easily decreases to below B_n . This leads to the expectation that the complete fusion cross-section should be very nearly the same as the fission cross-section and very few compound nuclei should survive the competition with fission.¹² It must be kept in mind that at each step in the evaporation process, i.e. every time a neutron is emitted, fission is competing. There results, in general, for high Z and A , a decrease in yield as more and more neutrons are emitted, i.e. $\sigma_{\max}(4n) > \sigma_{\max}(5n) > \text{etc.}$ The excitation functions for compound nucleus reactions exhibit a maximum cross-section with roughly a gaussian shape (Figure 1). At too low energies, not enough excitation energy is imparted to emit the desired number of

neutrons while at higher energies, too much excitation energy is present, resulting in the emission of additional neutrons. The energy of the maximum cross-section is in part a function of the Coulomb barrier and the neutron binding energy.

While the exact mechanisms for transfer reactions are not known, certain properties, characteristic of these reactions, serve to distinguish them from the compound nucleus mechanism.¹³⁻¹⁸ The differential cross-section for transfer products peaks at center of mass angles greater than 10° for bombarding energies near, but above the Coulomb barrier. The peaks of the excitation functions generally occur at higher energies, are more slowly decreasing or actually plateau at higher excitation energy, and at times have higher cross-sections than those for compound nucleus formation (for the same measured isotope). For the compound nucleus system, the range of the recoiling nuclei increases with increasing bombarding energy, while such a trend is not the case with transfer reactions. For example, Hahn et al.¹⁷ measured the recoil range of californium isotopes from $^{12}\text{C} + ^{239}\text{Pu}$ and found their ranges to vary from 1.6 times the expected complete fusion value at 67 MeV for this system down to 0.6 of the complete fusion value expected at 97 MeV. A characteristic similar to the compound nucleus mechanism is a general trend toward neutron deficient species resulting from neutron emission from the excited, newly-formed transfer product.¹³

One possible mechanism for these transfer reactions¹³⁻¹⁷ involves a two-step process. In the first step, a cluster or a single nucleon is transferred from the projectile to the target nucleus forming a recoil nucleus with a particular recoil velocity, $\vec{v}_{R_1}$, in the direction

dictated by the incoming projectile (0°). In the second step, scattering between the recoiling nucleus and the remaining light residue gives rise to a velocity, $\vec{v}_{R_2}$, and a scattering angle of ϕ for the recoil nucleus. The final velocity of the recoiling nucleus, $\vec{v}_f$, is the vector sum of $\vec{v}_{R_1}$ and $\vec{v}_{R_2}$. Using this model Hahn and co-workers¹⁷ were able to reproduce the observed experimental trends of recoil range, angular distribution, average recoil energies, and excitation energies of californium isotopes produced in the $^{12}\text{C} + ^{239}\text{Pu}$ reaction. A second possible mechanism proposed by Galin et al.¹⁹⁻²⁰ requires the transfer to occur at the point of closest approach, $R_{\min}$, i.e., the incoming and outgoing trajectories are expressed solely in terms of Rutherford scattering. Calculations based on this model were also consistent with the conclusion that the californium isotopes measured by Hahn et al. were produced via a transfer of ^8Be to the plutonium target although a number of details in the predictions of the two models are distinctly different.

Information on the mechanism of production is important in designing experiments to search for new elements and isotopes since quantitative information on the competing processes in heavy ion reactions can demonstrate which reactions are most favorable for production of a particular nuclear isotope.²¹ In addition, the distinct characteristic differences between compound nucleus and transfer reactions have been used to aid in the identification of heavy-ion reaction products.²² It should be noted, however, that there are some intervals of bombarding energy where recoil energies and angles from compound nuclei and transfer reactions are quite similar, and great care should thus be taken when

such properties are used to identify new isotopes.¹⁷

B. Purpose of this Study

It was decided to investigate the production of neutron deficient actinide isotopes with respect to these reactions. Recently, extensive efforts have concentrated on promising mechanisms for the production of superheavy elements.²³⁻³¹ Very little work, however, has concentrated on investigating the neutron deficient actinides. Berkelium, in particular, has been ignored. As can be seen from Figure 2 the light berkelium isotopes are "missing". Until the work presented in this thesis was completed, the lightest isotope of berkelium known was ^{243}Bk , the first berkelium isotope discovered.³² (There is some evidence for a spontaneously fissioning ^{242}Bk isomer.³³) Few studies have been made of the berkelium isotopes since the work of Chetham-Strode³⁴ in 1956. He investigated the decay properties of berkelium 244-249 and of californium 244-247.

This work has concentrated on studies of production of the light berkelium isotopes, ^{242}Bk in particular, and on the possibility of decay by spontaneous fission of light isotopes of plutonium, curium, and californium. It was hoped that through these investigations pertinent information would be gained concerning competition between fission and neutron emission, the amount of transfer versus compound nucleus reactions, and primary decay modes for neutron deficient actinide isotopes.

C. Characteristics of Light Berkelium Isotopes

The investigation of berkelium isotopes was undertaken for a number

of reasons. As has been pointed out, the light isotopes of berkelium are missing (Figure 2). Concern was expressed as to the reason for this "hole". Was it simply that no one had performed a conscientious search for these isotopes; or more interestingly, were they unusually short-lived and/or produced in unexpectedly small yield? Several years ago some preliminary searches for light isotopes of berkelium and curium were made, but to no avail.³⁵ While the parents of the berkelium isotopes 239, 241, and 242 are known, these daughter isotopes have not been observed.³⁶⁻³⁹ A spontaneously fissioning isomer of ^{242}Bk has been reported,³³ but methods of identification based solely on spontaneous fission decay are always subject to question.⁴⁰

An additionally attractive feature of berkelium is its chemistry. Because it exhibits a +4 oxidation state it can easily and quickly be removed from the +3 actinides allowing chemical separations to be performed on short-lived isotopes. The chemical separation of berkelium is extremely important both for the identification of the element itself as well as the identification of a new isotope (via the observation of the daughter of the new species in the parent chemical fraction).

Recent empirical calculations by Kolesnikov and Demin predict an electron capture half-life of 13 min and 27 min for berkelium 241 and 242 respectively, and alpha half-lives of 25 and 210 days.⁴¹ These long alpha half-lives correspond to α -branching ratios of 0.036% and 0.009% respectively. The empirically extrapolated alpha energies would be 7.1 MeV and 6.84 MeV. Keller and Müzel also predict similar half-lives.⁴² Since these berkeliums are predicted to decay primarily by electron capture, their identification had to be based on the characteristic

x-rays of their curium daughters. This necessitated a short, (due to short half-lives) clean chemical separation of berkelium from isotopes having X or gamma-rays near the K X-rays of curium. The chemical separations used are described in Section III.

There are no reported cross-sections for the production of neutron deficient berkelium isotopes by ions any heavier than alpha particles. The decision, then, as to what target/projectile combinations to use was based on the cross-section predictions of Alonso⁷⁴ described in Section V-D. Boron on uranium was chosen as the most suitable combination due to the large cross-sections predicted (see Figure 1), the high intensity boron beams available at the 88-inch cyclotron, and the ready availability of various uranium isotopes for targets. Nitrogen on thorium, although not as high in predicted cross-section, was another suitable combination. Alphas and protons were not considered as projectiles because they would not have supplied enough energy to allow the recoils to escape from the target, thus necessitating dissolution of the highly α -active, irradiated target.

D. Spontaneous Fission Studies

The spontaneous fission studies of plutonium, curium, and californium were undertaken to investigate the possibility of the existence of a spontaneous fission island in the very neutron deficient region of these actinides. ^{244}Fm , the lightest known fermium isotope, decays by spontaneous fission with a half-life of 3.3 msec. ^{232}Am , ^{234}Am , and $^{227/228}\text{Np}$ are also reported⁴³ to decay by spontaneous fission. Thus, there has been some speculation^{44a,b,c} that there may exist a region of spontaneous

fission activity in the neutron deficient area. Interest also exists in seeing how the spontaneous fission half-lives of these light actinides might compare to empirical extrapolations from the known isotopes.

The systematics of the spontaneous fission half-lives for even-even isotopes of elements from plutonium to nobelium are shown in Figure 3. Not only does the 152-neutron subshell produce a well-known effect on the spontaneous fission half-lives, but it also shows up as a characteristic kink in the alpha energy systematics with an alpha energy minimum around 152 neutrons (Figure 4). The dashed lines in Figure 3 show purely systematic predictions obtained by fitting a parabola of the form $\text{LOG } T_{1/2 \text{ SF}} = A + B(N - N_0)^2$ to known points for plutonium, curium, and californium. These predictions do not take into account the fact that the light isotopes, plutonium in particular, are expected to approach spherical shape as the 126-neutron subshell is approached and, therefore, may not have spontaneous fission half-lives as short as these predictions. It should be kept in mind, however, that the more neutron deficient an isotope is, the more β -unstable it is. It is quite possible that the effect of β -instability may completely override any contribution due to the 126-neutron subshell and these light isotopes may, indeed, have extremely short half-lives. The spontaneous fission experiments in this work were undertaken in an attempt to provide information on the above question and to investigate, more generally, the properties of the light actinides.

11. EXPERIMENTAL METHODS

A. Target Preparations

A number of techniques⁴⁵ were employed to prepare suitable targets for the various bombardments. These included electrodeposition, centrifugation, vacuum evaporation, and electrospraying. The particular technique used was determined by the target material itself, the amount of material available, the desired thickness and size of the target.

Electrodeposition techniques⁴⁶⁻⁵⁰ were used to make thin, uniform targets of uranium and lanthanide elements. Targets thicker than $\sim 500 \mu\text{g}/\text{cm}^2$, however, were very brittle and easily cracked and flaked off the target backing. Thick targets were made by plating the target material (lead or bismuth) directly onto the rotating drum used in the spontaneous fission experiments.

Targets of various heavy elements such as lead were prepared by centrifugation of a very fine sulfide precipitate in a special centrifuge cone which had the target backing mounted at the bottom of the cone. These targets were fairly thick ($\sim 1-2 \text{ mg}/\text{cm}^2$). They were, unfortunately, very fragile and could not generally be used more than a few times. For the spontaneous fission experiments, HgS was painted in a thin film directly onto the drum. Vacuum evaporation^{42,52,53} was a successful technique for nonradioactive targets and for those targets having a large supply of starting material available. This technique suffered from large contamination problems which also made recovery of unused starting material very difficult.

In an effort to overcome the various disadvantages of the techniques

mentioned above, another method of target preparation was developed and adapted for use with radioactive materials. The technique is known as electrospraying.⁵⁴⁻⁶¹ A detailed account of the methods used and developed can be found in reference 61, but a simple explanation follows.

A schematic of the apparatus is shown in Figure 5. The target material, in any soluble form - usually the nitrate form, was dissolved in acetone and pipetted into the glass capillary. A voltage of 4500-5000V, DC, was applied to the wire extending down the center of the capillary. The acetone solution formed a fine spray which was deposited on the target backing. The target was at ground potential and several hundred degrees, C°. Mica screens were used to collimate the spray and a certain amount of focusing of the spray was possible by applying a voltage to a metal ring set just in front of the target. The result was a uniform layer of target material in oxide form on the target backing. Once the target was finished, it was heated to red hot in an induction coil to insure all the target material was in the oxide form.

Using this procedure it was possible to produce targets ranging from a few $\mu\text{g}/\text{cm}^2$ to several mg/cm^2 . Beryllium was the typical target backing material although havar, dural, nickel, and aluminum have also been used. Electrospraying had advantages over a number of other target-making techniques in that the oxide targets prepared in this manner were extremely sturdy and uniform up to $5 \text{ mg}/\text{cm}^2$. Only small amounts of material were needed as the plating efficiency was quite high. The system was easily adapted for use in a glove box and thus radioactivity was not a problem. This method proved to be the most successful and reliable of those used. Consequently almost all targets used in this

study were prepared via the technique of electrospraying.

The typical target used in these studies was composed of the oxide form of the target material on 0.5 or 1.0 mil Be foil. The targets were 5/16 inches in diameter and approximately $400 \mu\text{g}/\text{cm}^2$ thick. Since a large number of experiments used a ^{235}U target, an analysis of the isotopic composition of the ^{235}U material (obtained from Oak Ridge National Laboratory) was performed using both α -counting techniques and mass spectrometry.⁶² The target material was determined to be $94.1 \pm 0.1\% \text{ } ^{235}\text{U}$ and $5.7 \pm 0.1\% \text{ } ^{238}\text{U}$.

B. Irradiation Techniques

1. Recoil Experiments

In an effort to minimize the radiation exposure to the chemist, it was decided to catch the recoiling compound nuclei (in experiments where chemistry was to be performed) on a thin catcher foil placed directly behind the target. This meant that only the catcher foil needed to be dissolved and the target could be reused any number of times. Further, the absence of macro amounts of target material greatly simplified the chemistry. The targets were usually thinner than the recoil range of the compound nuclei so that almost all the compound nuclei would escape. The catcher foil, on the other hand, was always a minimum of twice the recoil range of the compound nuclei to insure that all the recoiling compound nuclei were caught. The ranges were calculated from the tables of Northcliffe and Schilling.⁶³

All irradiations were carried out in the high level cave area of the 88-inch cyclotron at Lawrence Berkeley Laboratory. Figure 6 shows

a schematic of the experimental set-up. A 1/4-inch diameter, water-cooled collimator was located approximately 4.5 inches upstream from the target. Since this collimator was smaller than the target area, all beam measured in the Faraday cup passed through the target. The target was cooled with liquid nitrogen gas flowing between a beryllium window upstream and the target itself downstream. The catcher foil was placed ~0.5 cm from the target and was cooled with ~150 torr of helium. This pressure of helium resulted in less than 0.2 MeV loss of energy for the recoiling compound nuclei.⁶³ That this pressure, indeed, did not alter the collection of recoils was shown by running in a vacuum with the same results as with helium. On occasion, the catcher foil was moved back and a collimator placed in front of it. This geometry discriminated against strongly non-forward peaked (i.e. non-compound nucleus) reaction products such as fission fragments and transfer products. In addition, while thicker than the compound nuclei recoil range, the catcher foils were still thin enough to allow the more energetic fission fragments and transfer products to pass right through, thereby further discriminating against non-compound nucleus products.

The amount of beam hitting both the collimator and the target - which were electronically isolated from one another - was continuously measured. This enabled the cyclotron operators to optimize the beam by minimizing the collimator reading while maximizing the target reading. To avoid inaccurate beam readings, two rare-earth magnets were mounted in front of the target to keep secondary electrons generated from the target contained there, while at the same time steering away secondary electrons generated elsewhere.

A 3-phase, 60 Hz beam wobbler was installed about 5 meters upstream from the target (after the last quadrupole magnet). This device is simply the stator of an induction motor and moves the beam in a small circle at a 60-Hz rate with a loss in beam intensity of ~15%. The use of the wobbler prevented the narrowly-focused beam from depositing a large amount of energy in a small target area and thus helped avoid excessive heating of the target in any one spot.

The targets used routinely withstood steady currents of 3 - 5μA of ^{3}B for several hours. In a number of instances they withstood beam bursts in excess of 15 - 20μA, but generally it was considered unsafe to operate at currents higher than 5 - 8μA. An interlock system was used which automatically cut the RF power of the cyclotron if the beam current went higher than some preset current or if the target broke (measured as a pressure rise in the beam line). As an additional precaution to protect the cyclotron from possible radioactive contamination due to breakage of a radioactive target, a slammer-valve⁶⁴ was designed and installed ~9.5 meters upstream from the target. Even a slight pressure rise in the beam line caused this valve to close within 3 msec, isolating the cyclotron from the target.

The energy of the cyclotron beam was determined by the frequency and the magnetic field of the cyclotron using the following relation:

$$\omega = (\text{har}) 48.616 \sqrt{1 - \frac{1}{(1 + \frac{E}{mc^2})^2}}$$

where ω = cyclotron frequency in megacycles/sec, E = beam energy in MeV, har = harmonic #, mc^2 = A of beam $\times (931.478) - q \times (0.511)$, and

q = charge state of beam. This method was checked independently with a magnetic analyzer and found to be quite accurate. The final energy of the beam in the target was calculated from the electronic stopping power tables of Northcliffe and Schilling,⁶³ taking into account the beam window-havar, aluminum or beryllium; the nitrogen cooling gas; the target backing (beryllium) and the target material itself. The bombarding energy was altered by using various materials and thicknesses for the beam window. In none of the cases considered was the beam energy degraded more than 13 MeV. The energy loss in the target was typically 0.5 MeV.

2. Spontaneous Fission Experiments

The technique for detecting spontaneous fission events from the decay of compound nuclei entailed either 1) the collection of recoil atoms on the surface of a rotating drum in cases where a thin target was used; or 2) plating or coating the drum surface with target material which then acted as a thick target and a collector for the recoils. Surrounding and facing the drum surface were strips of muscovite mica which served to record the fission fragment tracks (Figure 7). The recoil atoms were caught on the drum and carried until they decayed, leaving tracks in the mica. These tracks were easily visible at 100X magnification after etching in hot hydrofluoric acid.

The targets were of two forms: thin (less than 1 mg/cm^2) targets on beryllium, aluminum, or havar backings or the drum itself was made to be the target by coating it with a thick (several mg/cm^2) layer of target material. The targets were prepared using the methods previously

described. Great care was taken to insure that the plating/painting was as uniform as possible.

All bombardments were carried out in the high level cave area of the 88-inch cyclotron. The targets were positioned perpendicular to the beam axis and the recoils passed in vacuum through a 20mm² opening to strike the drum surface (21mm from the target). In cases where a fissionable target was used, such as uranium, a very thin ($\sim 26\mu\text{g}/\text{cm}^2$) aluminum stopper foil was placed between the target and the drum. This prevented transfer of any target material to the drum which would create a spontaneous fission background. The thick drum targets were placed at an angle of 72° with respect to the beam axis to maximize the number of surface atoms bombarded.

The beam current was measured directly from the rotating drum which served as a Faraday cup by graphite brushes in contact with the rotating drum shaft. The drum was cooled by water flowing through the shaft. Drum speeds, which varied from 1 to ~ 2000 rpm, were measured stroboscopically from the frequency of interruption of a light beam by a perforated disc mounted on the drum shaft. Beam energies were determined as in the recoil method.

The stationary micas were mounted 0.8mm from the drum surface, in strips measuring 3 cm x 5 cm. There was a total of 14 micas which completely surrounded the drum surface except for the 2 cm gap at the beam window.

III. CHEMISTRY.

A. Chemical Properties of Berkelium

As do all the transuranium elements, berkelium exhibits the $+3$ oxidation state in solution; Bk(III) is carried by LaF_3 or $\text{La}(\text{OH})_3$ while berkelium nitrate, sulphate, chloride, sulfide and perchlorate are soluble in water. Bk(III) can be readily complexed with thenoyltrifluoroacetone (TTA) and extracted into benzene or xylene, and at pH 4 separation from curium and americium can be effected.⁶⁵ Moderately strong complexes can also be formed with citrate, lactate, tartrate, α -hydroxyisobutyrate, ethylene diamine tetraacetic acid (EDTA), acetate, and thiocyanate.

Bk(III) follows closely the ion exchange behavior of its lanthanide homologue - terbium. Figure 8 shows the rates of elution of californium, berkelium, curium and americium compared with their lanthanide homologues on Dowex-50 resin (cation exchange) and citrate eluant at 87°C .⁶⁶ A distinct break occurs between gadolinium and terbium and also between curium and berkelium as a consequence of the abrupt change in ionic radius which occurs upon completion of the $4f^7$ shell in gadolinium and the $5f^7$ shell in curium. This discontinuity in ionic radius is manifested as a significant change in complex ion formation resulting in a gap in the elution curves. Berkelium elutes ahead of the lanthanide elements on a saturated HCl-Dowex-50 column⁶⁷ indicating that berkelium forms a stronger chloride complex than do the lanthanides. Berkelium is also held, albeit weakly, on a Dowex-1 column (anion resin) in saturated HCl⁶⁸ demonstrating substantial formation of anionic chloride complexes.

Berkelium's lanthanide homologue, terbium, exhibits not only a $+3$ oxidation state, but (in the solid state) a $+4$ state as well ($4f^7$). Berkelium achieves the particularly stable half-filled $5f^7$ structure by also assuming a $+4$ oxidation state. Further, in general, higher oxidation states are more easily realized in actinide elements than in lanthanides; thus the $+4$ state of berkelium is stable in aqueous solution as well as in the solid phase. This is a particularly convenient property of berkelium because it allows Bk(IV) to be rapidly and quantitatively extracted into such reagents as bis(2-ethylhexyl orthophosphoric acid (HDEHP)/heptane from nitric acid leaving behind the $+3$ lanthanides and actinides.^{69,70} Bk(IV) can also be extracted quantitatively into TTA/xylene⁷¹ with excellent separation from alkalies, alkaline earths, trivalent lanthanides, Ru, Zr, Nb, U, Np, Pu, Am, Cm, Fe, Ni, Al, and Ag.

B. Chemical Properties of Americium and Curium

Americium is the actinide homologue of europium and most commonly exhibits the properties typical of a trivalent actinide or lanthanide. Other valence states, namely 0, IV, V, and VI have been found, but only Am (III), (V), and (VI) are stable in aqueous solution. The higher valence states may, at times, be used to effect chemical separations of americium from curium.

Curium is the actinide homologue of gadolinium and exhibits a single valence state of $+3$ in solution. Thus curium follows the expected behavior of trivalent lanthanides and actinides toward group reagents and is very similar to trivalent americium. Curium (III) and americium (III)

quantitatively precipitate as the fluoride, hydroxide, and oxalate. The $+3$ actinides tightly bind to cation exchange resin and moderately strong complexes are required to remove them. The $+3$ actinides elute ahead of the lanthanides from Dowex-50 resin in saturated HCl^{67,72} making possible an effective group separation of the actinides and lanthanides. Cm(III) may be separated from Am (III) on a Dowex-50 column at 87° with α -hydroxyisobutyrate.⁷³ There is little or no tendency for Am (III) or Cm (III) to absorb onto anion resin from 0.1M to concentrated H₂SO₄, HCl or HNO₃.^{68,74,75}

Both americium (III) and curium (III) can be extracted into TTA/benzene at pH > 3.⁶⁵ Goffart and Duyckaerts⁷⁶ have studied the extraction of curium (III) and americium (III) from nitric acid solutions by tri-n-octylphosphine oxide, TOPO, in benzene and also by tri-n-butylphosphine oxide, TBPO. Curium and americium may be separated by a solvent extraction procedure using tributyl phosphate, TBP, developed by Peppard and Gray.⁷⁷ Curium (III) is extracted to a greater extent than americium (III) and is concentrated in the organic (TBP) phase by use of counter-current methods, while americium (III) migrates to the aqueous (10-16M HNO₃) phase.

For a more detailed accounting of the chemistry of the actinide elements, the reader is referred to references 78-84 which are only a small sample of the multitude of publications concerned with actinide chemistry.

C. Separation Procedures

1. Berkelium

A variety of chemical separations of berkelium from the catcher foils was explored before the final chemistry was chosen. Speed was one of the primary considerations. Searching for half-lives less than 20 minutes (ultimately ≈ 7 min) ruled out the use of an α -hydroxy-isobutyrate/Dowex-50 column. In fact, the time element ruled out most of the more classic separation schemes aimed at isolating berkelium (III). Attention was therefore focused on the separation of berkelium (IV). Berkelium (IV) can easily be extracted away from $^{+3}$ actinides, $^{+3}$ lanthanides, Pu, U, Th, Al and a score of other elements in a very short time (< 5 min) compared to the more involved procedures necessary for berkelium (III) separation. The following procedures were all employed in the course of this work. The HDEHP solvent extraction procedure, however, was determined to be the most efficient and practical separation considering the various experimental constraints. It is important to note, however, that all three procedures gave consistent experimental results when corrected for chemical yield and the time from the end of bombardment. Additional procedures for the separation of berkelium (IV) may be found in references 85 - 91.

a. TTA Solvent Extractions

The procedure of F.L. Moore⁷¹ was adapted for speed and small volume work, but essentially remained unchanged (Figure 9). The Al catcher*

*NOTE: 99.999% pure aluminum was used to eliminate contaminating gamma rays resulting from products due to boron and nitrogen on Ni, Cu, Cr, etc.

was dissolved in a minimum amount of 7M HCl, dried, converted to nitrate form with 1 drop of 8N HNO₃ and dried again. 450 λ of 1.0 N HNO₃/0.2M Na₂Cr₂O₇ were added and the solution was heated for 1 minute. 50 λ of 5N H₂SO₄ were added and the solution cooled quickly in an ice bath. 500 λ of 0.5M TTA/xylene were added to the aqueous solution and the two phases were mixed using a Vortex mixer for ~1 minute. The aqueous phase was removed and the organic phase was washed with 500 λ of 1.0 N H₂SO₄/0.2M Na₂Cr₂O₇. The aqueous wash was removed and the organic phase was contacted with 250 λ of 10N HNO₃. The berkelium was found in this aqueous back-extraction phase. It was then plated on a platinum disc for α , γ , and/or X-ray counting. To further speed up this separation, the wash of the organic phase and the back extraction were eliminated and the volume of the organic phase was cut to 250 λ . The chemical yield after these modifications was ~50%. This particular separation is valuable because, unlike the separations based on HDEHP, this procedure effects a separation of berkelium (IV) from Ce (IV), Pu (IV), Np (IV) and U from aqueous solutions of HNO₃ and to a lesser degree from H₂SO₄, or HCl. Unfortunately, however, this chemistry extracts ²³³Pa which is produced in abundance from a ²³⁵U target and even worse, has a 111 KeV gamma ray which interferes with the observation of the Cm K α ₁, X-ray (109.29 KeV). Thus this chemistry was useful only for alpha analysis of the berkelium fraction.

b. HDEHP Solvent Extraction

A number of authors have reported procedures for the extraction of Bk(IV) by bis(2-ethylhexyl) orthophosphoric acid -- (C₈H₁₇O)₂PHOH,

HDEHP.^{70,80,81,92-95} Using this extractant under the appropriate conditions, tetra- and hexavalent lanthanide and actinide ions may be removed from an aqueous nitric acid phase into the HDEHP while the trivalent ions present remain preferentially in the aqueous phase. Thus this technique is not suitable for separating Bk (IV) from Ce (IV) or macro amounts of U (VI) or Pu (IV). On the tracer scale, however, uranium and plutonium were not significant problems.

The procedure of Peppard, Moline, and Mason⁹² was found to be quite efficient (60%) even after it was modified for time (all mixing or equilibration steps were cut to ~1 min) and volume (volumes used were on the order of 250 μ l). It had one major drawback in that the berkelium was oxidized by KBrO₃ in 10M HNO₃. KBrO₃ is unstable at such a high acid concentration requiring the KBrO₃/HNO₃ solutions to continually be freshly prepared. Further, the release of bromine gas from the solution required special, careful handling in fume hoods and created a generally "nasty" environment.

This problem was overcome in the separation procedure of Knauer and Weaver.⁷⁰ In this case, berkelium was oxidized with chromic acid (instead of KBrO₃) in 4M HNO₃. It was then extracted into 0.15M HDEHP/n-dodecane. The exact procedure adopted for this work, shown in Figure 10, is as follows:

- 1) The aluminum catcher foil was dissolved rapidly in ~1-2 drops of 2N HNO₃ in the presence of mercuric ion.

- 2) 500 μ l of 0.1M CrO₃/4N HNO₃ were added followed by 500 μ l of 0.15M HDEHP in dodecane. The dodecane had previously been scrubbed with

concentrated H_2SO_4 to destroy possible contaminants.

3) The phases were mixed for approximately 4 minutes -- the time it took for transportation of the solution from the cyclotron to the building where the chemistry was completed and the counting performed.

4) Upon arrival at the chemistry building, the phases were separated and 250 μ l of 1.5M H_2O_2 /10M HNO_3 were added to the organic phase.

5) After mixing for 1 minute, the aqueous phase was removed and either taken to dryness on a platinum disc for later alpha analysis or absorbed onto 1/4-inch diameter Whatman 41 filter paper discs for gamma analysis.

This procedure proved to be the fastest and most efficient chemistry and was therefore used in the majority of experiments. The average time from the end of bombardment to the detector was 12 minutes with a chemical efficiency of 50%.

c. HDEHP Column Chromatography

Before the final chemistry was chosen, a method for the extraction of berkelium (IV) by liquid-liquid chromatography was devised. It was felt that this form of extraction might be faster and/or more efficient than direct solvent extraction. Both beliefs proved true under appropriate conditions.

The columns (2.5 mm i.d.) consisted of HDEHP adsorbed on 60-70 mesh teflon powder (obtained from Ken Hulet at Lawrence Livermore Laboratory) as a solid support.⁹⁴ The HDEHP-teflon material was prepared by slurring 0.75 grams of teflon with 100 μ l of HDEHP and 10-15 ml of acetone

with the excess solvent being evaporated at room temperature. The columns were dry-packed to a height of ~5 cm by gently packing the HDEHP/teflon material into the column with a glass rod. Prior to packing, the columns were made hydrophobic by exposure to dichlorodimethyl silane vapors for at least 24 hours.

After packing, a minimum of 10 column volumes of 0.1M $\text{CrO}_3/4\text{M HNO}_3$ was run through the column to prepare it for the berkelium extraction. The dissolved catcher foil was loaded in a minimum volume of 0.1M $\text{CrO}_3/4\text{M HNO}_3$. Pressure was applied to obtain a flow rate equal to 30 drops/minute. Upon addition of the eluant, 1.0M $\text{H}_2\text{O}_2/10\text{M H}_2\text{O}_2$, the column turned from orange to green and a significant number of gas pockets were formed in the column bed. A minimum of 5 column volumes was necessary to remove the berkelium. In spite of the rapid flow rate and disrupted column bed, the chemical yield was typically 70%. The time for this chemistry was actually shorter than the solvent extraction procedure when both were performed side by side in the laboratory. However, since the solvent extraction method could be carried out during transportation to the chemistry building, the total time from the end of bombardment to the detector for the solvent extraction method was shorter. In addition, the columns could not be reused and had to be cleaned and repacked with fresh material. Thus this procedure was extremely inconvenient for bombardments in which a number of catcher foils were processed in a short period of time. However, it was the best procedure used both in chemical yield and time for chemistry. Only the constraints due to having to transport the catcher foils to another building resulted in the solvent extraction method being

avored.

A note about the purification of the HDEHP is appropriate at this point. Typically, HDEHP is commercially provided as a mixture of the di-ester (used in this work) along with up to several percent of mono-2-ethylhexyl orthophosphoric acid, polyphosphates, pyrophosphates, 2-ethylhexanol, other alcohols, and frequently various metal ions such as iron (III). The monophosphates and the pyrophosphates are the most active impurities. They require careful removal, as even a trace of these impurities greatly increases the distribution coefficients of many elements.

Two sources of HDEHP were used in this work:

1) Several milliliters of the di-ester, purified using the techniques of Stewart and Crandall⁹⁶ and Peppard et al.,⁹⁷ were obtained from Ron Loughheed at Lawrence Livermore Laboratory. The original HDEHP was from Eastman Kodak Co.

2) HDEHP, 96% di-ester, was also obtained from ICN-K&K Laboratories, Plainview, NY. Purification of separate aliquots was carried out using the above techniques^{96,97} on one aliquot and the alkali washing technique of Gureev et al.⁹³ on another. Since the chemical separation results from all three "batches" were consistent, the procedure of Gureev et al. was preferred since it was simplest and required the least volume of reagents. For even greater purification, the procedure of McDowell et al. is recommended.⁹⁸

2. Americium and Curium

The flow chart in Figure 10 demonstrates the procedure followed

for the separation of curium/ameridium fractions from the catcher foils and from the parent berkelium fraction. This chemistry is a classical transplutonium chemistry based on procedures given in references 81 and 99.

The sample was dissolved in 9N HCl and a few drops of water added. After the addition of ~250µg of lanthanum carrier, 100λ of concentrated hydrofluoric acid were added. After stirring, the solution was allowed to stand for several minutes before centrifuging. The supernatant was removed and the precipitate was washed several times with 2N HNO₃/2N HF prior to being dissolved in 2 drops of saturated boric acid and 150λ of concentrated HNO₃. This step effectively separated the ⁺³ actinides and lanthanides from many contaminating ions of other valences. Thorium, uranium, and plutonium were also precipitated and were separated later on an anion exchange column.

The lanthanides and actinides were removed from the boric acid solution by precipitating them as hydroxides. To the above solution of boric acid/HNO₃/lanthanides/actinides were added ~12 drops of concentrated NH₄OH. After centrifuging and removing the supernatant, the precipitate was washed 3-4 times with water before redissolving it in a minimum volume of 8N HNO₃. Thorium, uranium, and plutonium were separated by loading the solution on a 2.5 cm x 2.5 mm i.d. Dowex 1x8 anion exchange column. Dry N₂ was used to force 8N HNO₃ through the column at a rate of 4 drops/min. The lanthanides and actinides were collected in drops 4-20 while plutonium, thorium, and uranium remained on the column. This solution was taken to dryness, time permitting, or else another hydroxide precipitation was performed as the volume reduction step. To the dried La/Ac fraction (or to the

hydroxide precipitate) was added ~1 drop of saturated HCl followed by ~1 mg of $\text{Ba}(\text{NO}_3)_2$. (Radium precipitated with the barium chloride.) The supernatant was removed and loaded onto a saturated HCl MP-50 column, 5 cm x 2.5 mm i.d., having a flow rate of less than 3 drops/min. The actinide elements eluted ahead of the lanthanides and were collected in drops 5-35. For a final purification, this solution was taken to dryness, redissolved in 1.5M HCl and loaded onto another Dowex 1x8 column, 2.5 cm x 2.5 mm i.d., with a flow rate of ~6 drops/min. This step removed lead and bismuth daughter products. Drops 4-20 were collected and taken to dryness on a 1-inch platinum disc for alpha and/or gamma counting. If this last fraction was contaminated with salts, it was taken to dryness, redissolved in acetate buffer ($\text{pH} \approx 4.5$) and contacted with an equal volume of 0.2M TTA/benzene several times.¹⁰⁰ The TTA fractions were combined and taken to dryness on platinum. After flaming the platinum disc, the result was a weightless sample ideal for α -counting. The chemical yield was typically 75% but was always measured directly with the use of ^{241}Am and ^{244}Cm tracers. The time for the entire procedure was approximately 100 minutes.

IV. COUNTING TECHNIQUES

A. Alpha Spectroscopy

Alpha spectroscopy¹⁰¹ of the samples was carried out using four gold-plated surface barrier detectors obtained from Ortec, Inc. The detectors had an average active area of 300 mm^2 , an energy resolution (FWHM) of 20 keV measured from the 5.485 MeV α peak of ^{241}Am , a noise width no greater than 15 keV (FWHM), a sensitive thickness of ~ 100 microns, and operated at a voltage of 50-100 volts.

The voltage for each detector was supplied through a pre-amplifier which also fed the output signal to a TranLamp-linear amplifier for alpha detection and to another linear amplifier for spontaneous fission detection (an option not used in these studies). All four signals from the linear amplifiers went into a router/mixer. From here, one signal at a time went to a biased amplifier which served to set the window size for alpha detection and the upper and lower window limits (in MeV). The signal was then fed into a pulse-height analyzer system consisting of a Texas Instruments 960A minicomputer, a Northern Scientific NS-621 analog-to-digital converter, 4096 memory storage locations (1024 channels/detector) and an Ampex magnetic tape deck. The data from each detector were stored in 1024 channels, and provisions existed for separate tag-words for each detector. Since all four detectors operated through the same biased amplifier and the same clock, they all were set for identical windows and counting intervals. The minicomputer offered a variety of options including integrating counts in variable channel intervals; recording the tag word, start time, stop time, live time, and counting

interval on magnetic tape; finding peak centroids; and printing-out the above information on a teletype.

The detectors were calibrated using thin ^{241}Am and ^{233}U sources in conjunction with a Porpul II B pulser. Least squares analysis of ~ 10 points between 4.0 and 9.0 MeV gave calibrations with fit parameters ≈ 0.99 (1.000 being a "perfect" fit). The calibrations were shown to be constant over periods of several weeks and varied no more than a few channels over longer periods. The detectors used were brand new at the start of these experiments and had essentially zero background over counting intervals of up to ten days.

B. X-Ray Spectroscopy

An intrinsic germanium solid-state detector (2 cm wide x 3 mm thick) with ~ 10 mil beryllium window was used in analysis of X-ray peaks. The detector was operated in conjunction with a 1024 channel (storage location) Northern Scientific NS-610 multi-channel analyzer, a Northern Scientific NS-400 data processor, and a NS-406M magnetic tape control. The system was calibrated using several isotopic standards: ^{241}Am (59.57 keV), ^{207}Bi (72.79, 74.96, 84.9, 87.4 keV), and ^{57}Co (122.1 keV). The energy resolution was 600 eV.

The absolute efficiency was not determined as this method of detection was used only to confirm the identification of the curium x-rays and to insure that there were no other conflicting gamma rays present in the spectra. The samples were, however, counted at constant geometry - approximately 0.5 cm from the beryllium window - allowing half-life measurements, but not absolute cross-section measurements to be made.

C. Gamma-Ray Spectroscopy

A 60-cm³ volume ORTEC coaxial-germanium, lithium drifted, diode detector was used in the gamma and the majority of the X-ray analyses. The detector was shielded by 5 cm of lead, 0.3 cm of steel and 0.3 cm of aluminum to reduce the contribution of background radiation and fluorescence induced in the lead. The detector and pre-amplifier were coupled to a linear amplifier then to a pulse-height analysis system consisting of a Northern Scientific NS-623 analog-to-digital converter, a Texas Instrument 960A minicomputer, 4096 channels for memory storage, and an Ampex magnetic tape unit. This system had similar capabilities to the alpha counting system, i.e. storage of tag-words, counting intervals, start, stop, and live time on magnetic tape, peak integrations, finding peak centroids, etc.

Energy calibrations and detector efficiencies were measured using a National Bureau of Standards Mixed Radionuclide Gamma-Ray Emission-Rate Point-Source Standard (SRM 4216-B) which contained the isotopes listed in Table 1. Least square fits of the channel versus energy data from this calibration source led to fit parameters, as in the alpha system, of 0.99 (1.000 being a perfect fit). The electronics were extremely stable with a calibration shift of no more than ~1 channel in several months. The resolution of the curium $K_{\alpha 1}$ X-ray peak (109.27 keV)¹⁰² was 1.5 keV FWHM. The samples were mounted on 8.8 cm x 6.2 counting cards and covered with a layer of saran wrap to prevent possible contamination of the detectors. All samples were counted 0.5 cm from the detector face. In this position the efficiency for 110-keV was 15%. The samples were only slightly active so the dead time was essentially

0%. The efficiency as a function of energy was previously determined for each shelf location¹⁰³ using the procedure of reference 104. A sample spectrum is given in Figure 15.

Table 1.

Energy Calibration Standards

<u>Nuclide</u>	<u>Energy (keV)</u>	<u>Half-Life</u>
¹⁰⁹ Cd	88.03	453d
⁵⁷ Co	122.06 136.47	270d
¹³⁹ Ce	165.8	137d
²⁰³ Hg	279.21	36.7d
¹¹³ Sn	391.7	115d
⁸⁵ Sr	513.98	65.2d
¹³⁷ Cs	661.64	30.1yr
⁸⁸ Y ^(d)	898.0 1836.1	106d
⁶⁰ Co	1173.21 1332.48	5.26yr

Background counts were taken to determine which peaks would be seen regardless of the sample being counted. ⁴⁰K along with the natural products of the uranium and thorium decay chains were the only major background activities noted in several days of counting.

D. Detection of Spontaneous Fission Events

Heavily ionizing, charged particles that pass through most dielectric (insulating) solids leave narrow tracks of intense damage.¹⁰⁵⁻¹⁰⁹ Since this damage is on the atomic scale, it must be amplified before it can be

observed. Often this is accomplished by chemical etching of the solid,¹¹⁰ a relatively simple technique. The chemical etchant preferentially attacks the damaged area while less rapidly attacking the undamaged material, resulting in an enlarging of the damaged area. It is then possible to observe these holes with an optical microscope (Figure 12).

Three basic types of solids are readily available for use as particle detectors: minerals, glasses, and organic polymers. For this work, flexible detectors were necessary to facilitate mounting around the drum and later mounting on glass slides for scanning. Thin mica sheets and the various organic polymers were especially suited to this purpose. In some preliminary experiments both lexan polycarbonate obtained from P. Buford Price (LBL) and mica detectors were used in an effort to compare them both for sensitivity, ease of handling and scanning. Mica was chosen because of its insensitivity to low Z particles, in contrast to lexan which was sensitive even to alpha particles at 0.3 MeV (reference 111, p. 18, Fig. 1-8).

The mica detectors were split and cut from 4-inch x 6-inch muscovite sheets, ASTM-R.¹¹² Muscovite, $KAl_3Si_3O_{10}(OH)_2$, is the best of all micas in dielectric strength, transparency, and perfection of cleavage. After cutting, the micas were annealed at $500 \pm 25^\circ\text{C}$ for 4 weeks.^{111,113} In addition to annealing, it was also necessary to pre-etch the micas for 4 hours in concentrated hydrofluoric acid at 60°C followed by 1/2 hr rinse in distilled water and a rinse with absolute ethyl alcohol. This preparation procedure eliminated any background fission tracks existing in the micas prior to irradiation. After a bombardment, the micas were masked and individually exposed to a nontransferable ^{252}Cf source to provide

a check on mica sensitivity and scanning efficiency. They were then etched for 1 hour in concentrated HF at 60°C, rinsed in water and dried with EtOH. For scanning purposes, the micas were mounted on 2 x 3 inch glass slides and examined at 100X magnification (Figure 12). The positions of the fission tracks were permanently recorded by an X-Y plotter interfaced to the microscope slide. Scanning efficiency was 100%.

As a check against tracks originating from fissionable material in the mica (due to the high neutron flux during the irradiation), the back as well as the front side of each mica was scanned. Corrections were made, when necessary, for any background tracks detected. With the particular batch of micas used, the average number of background tracks/mica was ~ 1 . To be sure that deeply buried, unannealed background tracks were not exposed on the final etch (to look like experimental tracks), 10 micas were annealed, pre-etched, scanned front and back, etched for an additional hour and scanned again. Not a single "experimental" fission track was found.

In determining the total detection efficiency of the system, account was taken of the emissivity of the drum (probability of the escape of one fission fragment from the decay of an atom - 88%), loss in the stopper foil ($\sim 4\%$), loss due to the 2 cm^2 collimator ($\sim 4\%$) and decay in the window for very short-lived activities. These figures were computed from particle range energy relations at specific trajectories.¹⁹ As a further check of the system, ^{244}Fm was produced in the reaction $^{233}\text{U}(^{16}\text{O}, 5n)^{244}\text{Fm}$ with a cross-section in excellent agreement with that previously reported by Nurmi et al.¹¹⁴

V. DATA REDUCTION AND ANALYSIS

A. Alpha Spectra

Since the samples counted were thin sources, the α -peaks seen were well-separated and well-defined as shown in Figure 13. The counts in each peak (identified by chemical fraction, energy, and half-life, if possible) were integrated, an A_0 determined, and finally a cross-section for production was calculated. This calculation included corrections for chemical yield, length of bombardment, detector efficiency, branching ratio of the particular alpha, etc. Chemical yields were determined by adding known count rates of ^{244}Cm and ^{241}Am to the samples before chemistry was begun.

B. X- and Gamma-Ray Spectra

1. Berkelium Samples

Identification of the berkelium isotopes was based on the observance of the K_{α_1} (109.27 keV) and K_{α_2} (104.59 keV)¹⁰² X-rays of curium and the half-lives obtained from their decay. In cases where the yield of berkelium isotopes was especially good, the K_{β_1} (123.41 keV) and K_{β_2} (122.29 keV) curium X-rays were also seen. Figure 14 is an example of a standard gamma-ray spectrum.

Since these X-rays occur in a region of high Compton and β^- background, i.e. in the region less than ~ 200 keV, careful attention was paid to correcting the observed count rates for a decaying background. This was accomplished using a program written for a Texas Instruments desk calculator by Walter Loveland at Oregon State University. Basically, average backgrounds were calculated on the

left and right-hand sides of the peak which were then used to calculate a background for the peak itself. A percentage error was also calculated to give an indication of the reliability of the results. This method proved to be highly satisfactory and gave quite reproducible results.

Once the decay rates were determined for both the K_{α_1} and K_{α_2} peaks in each spectrum, half-life analyses were carried out using several methods. Two computer codes, CLSQ¹¹⁵ and FRANTIC,^{116a,b} were employed. Due to the poor statistics from the low count rates, however, both these programs failed to resolve the ^{243}Bk background. A more successful approach was reached by using a desk calculator for least square fitting and subtracting the long-lived components from the shorter components. While taking longer to complete, this method of "hand" analysis proved superior to existing computer codes since it could be carried out even with very low statistics. Once half-lives were established, it was a simple matter to determine A_0 values.

A number of considerations went into the cross-section calculations. Besides the obvious corrections for beam level, target thickness, detector efficiency, bombardment duration and chemical yield (determined by the growth of ^{249}Cf from ^{249}Bk tracer) a correction also needed to be applied to the A_0 value. The A_0 values obtained were the sum of the K_{α_1} and K_{α_2} curium X-rays. A correction had to be applied 1) to account for those K X-rays not analyzed and; 2) to account for the fluorescence yield (w_K) of K X-rays from the curium decay. Dittner and Bemis¹⁰² experimentally determined the relative transition probabilities of K X-rays for curium which were found to be in excellent agreement with predicted values of Lu, Malik, and Carlson.¹¹⁷ The experimental values

were used to correct for the missing $K_{\beta 1}$, $K_{\beta 2}$, $K_{\beta 3}$, etc. X-rays.

An estimate for the fluorescence yield of K X-rays from curium taken from Fink et al.¹¹⁸ was 97%.

2. Americium/Curium Samples

A rather different approach was used in the analysis of data where the statistics were more reasonable, for example, in the cases involving americium/curium samples. Here a peak finding program, SAMPO,¹⁰⁴ was used to locate photopeaks, calculate their energy, and measure their area. From the SAMPO analysis, the peaks were identified on the basis of half-life, energy, and the presence of sister peaks. The primary source for the peak identification data was the recent summary of gamma-ray information by Binder¹¹⁹ et al. Cross-sections were then calculated considering the percentage of the gamma-ray, length of bombardment, beam level, chemical yield, etc.

C. Spontaneous Fission Data

From the positions of the fission tracks, as recorded by the X-Y plotter interfaced to the scanning microscope, plots were made of the number of tracks versus time (position). The time intervals were calculated from the circumference of the drum and its revolution speed. In this way half-lives could be determined for observed fission activities with half-lives within a range which depended upon the drum speed; that is, if the drum rotated very fast, then a spontaneous fission activity with a long half-life would have appeared as a uniform background activity whereas if the drum rotated too slowly, a short-lived activity would have decayed before reaching the mica detectors. In cases where no

activity greater than background (< 1 track/mica) was seen, only half-lives between the limits due to the drum speed could be set. The activity would have to be very short- or very long-lived not to be seen.

Cross-section calculations were corrected for total efficiency (see Section IV-D), beam intensity, length of bombardment, etc. In cases where no activities were observed only upper limits were obtained.

It must be kept in mind that the observance of a spontaneous fission activity in mica track detectors yields nothing more than half-life information. There is no mass nor Z identification and, as will be shown later, simply assuming $Z_1 + Z_2 = Z_{1+2}$ is not a sufficient determination of the Z of the activity.

Estimates of limits to spontaneous fission branching ratios and thus of spontaneous fission half-lives were obtained by assuming production cross-sections and half-lives for the activities, calculating the spontaneous fission branch ($\frac{\sigma_{SF}}{\sigma_{PRED}} \rightarrow SF$) and from that, the spontaneous fission half-life limit ($\frac{t_{1/2}^{pred}}{SF \text{ branch}} < t_{1/2}^{SF}$). The half-lives used for these calculations were obtained from the empirical estimates of alpha and electron capture half-lives of Kolesnikov and Demin,⁴¹ of Taagepera and Nurmia,¹²⁰ from the methods of Viola and Seaborg,¹²¹ and of Keller and Müzel.⁴² The margin for error here is large, but should certainly be less than a factor of 5-10. The more dramatic errors are likely to be due to the assumed cross-sections. The cross-sections actually used were reduced by a factor of ~ 15 from the calculated results (JORPLE).¹²² The basis for the cross-section calculations and the

rationalization for this reduction in cross-section will be explained in the next section.

D. Cross-Section Estimates

Estimates of the cross-sections for the formation of compound nuclei in heavy ion reactions are extremely helpful to the experimentalist. They enable choices to be made regarding which target/projectile combinations to use, what yields to expect, what beam energies are optimum, etc. for the production of a particular isotope. These estimates can be particularly useful when there are little or no experimental data in the region of interest, as in the case of neutron deficient actinide isotopes.

The cross-section calculation code employed, JORPLE, developed by Jose Alonso^{122,123} and J.O. Rasmussen^{123,124} and based on a computer code of Sikkeland and Lebeck¹²⁵, considers the formation of the compound nucleus followed by competition between fission and neutron emission as the major modes of deexcitation. Consideration is given to the interaction potential, which is analogous to that of Thomas¹²⁶ and of Rasmussen et al.,^{123,124} to angular momentum effects, to nuclear deformation, to fusion probability using the Hill-Wheeler transmission formula,¹²⁷ to compound nucleus formation using the formula of Blatt and Weisskopf,¹²⁸ and finally to deexcitation of the compound nucleus by neutron evaporation which is based on the Jackson model,¹²⁹ and corrected for fission competition using the empirical formula of Sikkeland¹³⁰ for Γ_n/Γ_f , the ratio of neutron emission to fission. A detailed explanation of the code may be found in the above references,

especially 122, 123.

As in all calculations of this sort, the cross-sections obtained are quite sensitive to the parameters used such as r_0 , d (diffuseness), nuclear shape and Γ_n/Γ_f . The parameters employed in the calculations gave the best overall fits to the experimental data through 1973 for heavy ion reactions for ions up to neon in this region of the table of isotopes. This does not mean, however, that these parameters give best fits in isolated regions such as on the very neutron deficient side of β -stability. Further, no consideration is given to evaporation of particles other than neutrons, although it is generally assumed that neutron emission is greatly favored over that of charged particles in this Z region. Neither are considerations applied to particular entrance channel problems of admittedly, perhaps, only a few specific projectiles, such as the break-up or dissociation of the projectile. Because these corrections are, in general, of little or no consequence, they are ignored. On the other hand, as in the case of the parameter values in isolated or local cases, they may indeed become important. The excitation functions calculated from this code agree extremely well in shape and energy with most of the experimental data in the actinide region. Unfortunately, it was discovered in this work that there are large discrepancies in cross-sections in the region of neutron deficient actinide isotopes as will be discussed at a later point. Figure 1 shows the calculated excitation function for the $^{11}\text{B} + ^{235}\text{U}$ system.

VI. EXPERIMENTS AND RESULTS

A. ^{242}Bk

Table 2 lists the various target/projectile combinations and the beam energies used to produce ^{242}Bk as well as the (H.I.,pxn) and (H.I., α xn) products, curium and americium, respectively. Each of the reactions used to produce ^{242}Bk was run at least two times. This was easily possible since the bombardments to produce this isotope were only ~25 minutes long. As many as 12 bombardments were carried out in one 8-hr shift. The experiments to produce the pxn and α xn products were each run once for 5-6 hours in an effort to produce as much activity as possible.

Table 2

Summary of Bombardments

<u>System</u>	<u>Reaction</u>	<u>Lab Energy (MeV)</u>	<u>Product(s)</u>
$^{10}\text{B} + ^{238}\text{U}$	5n,4n	60	$^{243}, ^{244}\text{Bk}$
$^{10}\text{B} + ^{238}\text{U}$	6n	67	^{242}Bk
	p7,5n	67	$^{240}, ^{242}\text{Cm}$
$^{10}\text{B} + ^{235}\text{U}$	4n	52-60	none observed
$^{11}\text{B} + ^{235}\text{U}$	4n	55-65	^{242}Bk
	4n	60	milked ^{242}Cm
	p5,3n	60	$^{240}, ^{242}\text{Cm}$
	α 4,3,2n	60	$^{238}, ^{239}, ^{240}\text{Am}$
$^{14}\text{N} + ^{232}\text{Th}$	4n	77	^{242}Bk
	p5,3n	77	$^{240}, ^{242}\text{Cm}$
	α 2n	77	^{240}Am
$^{15}\text{N} + ^{232}\text{Th}$	5n	76-93	^{242}Bk

The first reaction, $^{238}\text{U}(^{10}\text{B}, 4-5\text{n})^{244, 243}\text{Bk}$, was a test run for the irradiation method, the counting system, and the chemistry. The energy was chosen to be close to the peak for the 5n reaction to produce ^{243}Bk . Since the energy used was actually on the low side of the 5n peak, a small amount of ^{244}Bk was also produced. Since the predominant gammas emitted by ^{243}Bk and ^{244}Bk occur at energies greater than 200 KeV, their detection efficiency is much lower than that for the curium X-rays. Further, the absolute intensities are not known. Thus since ^{243}Bk - and ^{244}Bk have half-lives very close to one another, only a combined cross-section of $14\mu\text{b}$ could be obtained from this experiment. This cross-section value is 30 times lower than that predicted by JORPLE.

After double-checking the irradiation system and the chemistry to be sure the low cross-section was not due to experimental error, work then advanced to the production of ^{242}Bk . Evidence for a short-lived (< 10 min) component in the curium $K_{\alpha 1}$ and $K_{\alpha 2}$ X-rays was found in the first experiment, $^{235}\text{U}(^{11}\text{B}, \text{xn})^{246-\text{x}}\text{Bk}$, even though the time from the end of bombardment to the detector was ~ 25 minutes. Subsequent bombardments using a shorter chemistry confirmed this component as having a half-life equal to 7.0 ± 1.3 m. A typical decay curve is shown in Figure 16. The short-lived component stands out quite strongly and is easily resolved from the longer-lived activity of ^{243}Bk (half-life = 4.5 hr).

The excitation function for this activity from $^{11}\text{B} + ^{235}\text{U}$ shown in Figure 17 agrees quite well in peak energy and width ($\text{FWHM} \approx 5$ MeV) with the Alonso calculations for the 4n reaction. The experimental cross-section of $\approx 10 \pm 2\mu\text{b}$, however, is approximately a factor of 20 below that calculated.

Since the peak energies for the 4n and 5n reactions are calculated to fall within approximately 5 MeV, it was unclear whether this activity was due to ^{241}Bk , ^{242}Bk , or both. An excitation function was, therefore, measured for any short-lived (half-life $\geq 3\text{m}$) component in the curium X-rays produced by ^{10}B on ^{235}U . No evidence for a short-lived activity was found over the energy range considered (52-60 MeV). This energy range should have maximized production of ^{241}Bk while that of ^{242}Bk should have been at least an order of magnitude lower.

As a further check on the mass number and the cross-section for production of this 7-minute activity, ^{232}Th was bombarded with ^{15}N . The measured excitation function is shown in Figure 18. Once more, the experimental FWHM (10.1 MeV) and peak energy agree well with the calculations for the 5n reaction, but the cross-section ($9 \pm 1\mu\text{b}$) is again ~ 25 times lower than predictions.

One of the best ways to confirm the mass number (and Z) of an isotope is to positively identify its daughter in the original parent chemical fraction. While long-term alpha counting of the various berkelium fractions gave hints (i.e. 7 counts expected in 4 days, 6 seen) of the presence of ^{242}Cm , the electron capture daughter of ^{242}Bk , it was decided to milk a number of separate bombardments for the ^{242}Cm daughter. A total of twelve 25-minute bombardments was carried out using ^{11}B on ^{235}U at 59.5 MeV. Berkelium was immediately separated (~ 4.5 minutes from EOB) from the dissolved foil, and the final berkelium fractions from each run were pooled. A day or so later, ^{244}Cm tracer was added and a curium fraction was removed from this combined berkelium fraction. Since the

half-life of ^{242}Cm is 163 days, it was necessary to count for long periods of time. Figure 13 shows a 10-day α -spectrum of the final curium fraction. The ^{242}Cm peak stands out quite nicely as does the ^{244}Cm . The ^{241}Am was used to calibrate the columns. From the amount of ^{242}Cm present, a cross-section for ^{242}Bk of $9.3 \pm 1.5 \mu\text{b}$ was found, in excellent agreement with the direct measurement of $10 \pm 2 \mu\text{b}$.

It was conceivable that the low cross-sections could be due to the decay of ^{242}Bk by alpha emission or even spontaneous fission. A number of berkelium fractions were alpha counted within 8 minutes for any short-lived activities. None were observed. In fact, no alphas at all were observed in the first few hours after bombardment! In order to avoid any losses due to chemical yield and time, a number of catcher foils were gamma and alpha counted approximately one minute from the end of bombardment (EOB). As in the chemistry runs, no (unidentified) alphas were observed. [The main alpha peaks seen were due to ^{211}At , ^{211}Po (from EC decay of ^{211}At), ^{213}Fr and ^{226}Th .] No information was gained from direct gamma counting of the catcher foils due to the intense β -, X-, and γ -ray background in the foil.

That the light berkelium isotopes do not decay by spontaneous fission has been shown by L.P. Somerville.¹³¹ He bombarded uranium isotopes with ^{10}B and ^{11}B at various energies as part of a study of delayed fission. No unaccounted for fission activity was observed resulting in a spontaneous fission branch $< 0.03\%$ for ^{242}Bk . Eastham and Grant¹³² bombarded ^{232}Th with ^{14}N at the peak for the $4n$ reaction and set a cross-section upper limit for the spontaneous fission of ^{242}Bk at 50nb for a half-life greater than 2ns .

B. (H.I.,pxn) and (H.I.,αxn) Data

Since it now appeared that the cross-sections for the compound nucleus, xn reactions, were indeed much lower than expected, the question was raised as to what products were being produced. It was decided to examine the pxn and αxn products, curium and americium respectively, produced directly in the same reactions and at the same energies used to produce ^{242}Bk . A single bombardment resulting in 16.9 μA-hrs of $^{11}\text{B}+3$ on ^{235}U was carried out. Chemistry was performed to separate curium and americium from the catcher foil. The resulting Cm/Am fraction was both gamma- and alpha-analyzed.

The results obtained are listed in Table 3. The berkelium data were not determined in the same bombardment as the curium and americium but are given for comparison. After correcting the total yield of ^{242}Cm for the amount of ^{242}Bk which decayed to ^{242}Cm , there remains a considerable yield of ^{242}Cm from other than compound nucleus mechanisms. The total contribution of ^{242}Am decay should be very small since the emission or transfer of only one alpha particle and no neutrons or other particles has a very small probability, especially at an excitation energy of ~43 MeV.

Data were also obtained from $^{10}\text{B} + ^{238}\text{U}$, $^{14}\text{N} + ^{232}\text{Th}$, and $^{15}\text{N} + ^{235}\text{U}$ as shown in Table 3. Only pxn information was gained in the ^{10}B case since the α(1-3)n products were too long-lived to be seen. The cross-section for ^{242}Bk from $^{10}\text{B}(^{238}\text{U},6n)^{242}\text{Bk}$ was found to be 8.9 ± 2.0 μb and from $^{14}\text{N}(^{232}\text{Th},4n)^{242}\text{Bk}$ to be 480 nb. Only an upper limit to the compound nucleus cross-section for ^{246}Es was determined due to its small cross-section and short half-life (7.3 m).

Table 3.

Yield of fission and α n Products

System	Energy	Product	Reaction	Cross-section
$11_B + {}^{235}U$	60 MeV	${}^{242}Bk$	4n	10 μ b
		${}^{243}Bk$	3n	1 μ b
		${}^{242}Cm$	p3n; ${}^{242}Bk \xrightarrow{EC}$	78 μ b
			${}^{242}Am \xrightarrow{\beta^-}$	
			p3n; ${}^{242}Am \xrightarrow{\beta^-}$	69 μ b
		${}^{240}Cm$	p5n	0.035 μ b
		${}^{240}Am$	α 2n	1500 μ b
		${}^{239}Am$	α 3n	410 μ b
		${}^{238}Am$	α 4n	< 1.6 μ b
$10_B + {}^{238}U$	67 MeV	${}^{242}Bk$	6n	8.9 μ b
		${}^{242}Cm$	p5n; ${}^{242}Bk \xrightarrow{EC}$	60 μ b
			${}^{242}Am \xrightarrow{\beta^-}$	
			p5n; ${}^{242}Am \xrightarrow{\beta^-}$	51 μ b
		${}^{240}Cm$	p7n	0.12 μ b
$14_N + {}^{232}Th$	77 MeV	${}^{242}Bk$	4n	0.480 μ b
		${}^{242}Cm$	p3n; ${}^{242}Bk \xrightarrow{EC}$	1.00 μ b
			${}^{242}Am \xrightarrow{\beta^-}$	
			p3n; ${}^{242}Am \xrightarrow{\beta^-}$	0.51 μ b
		${}^{240}Cm$	p5n	0.055 μ b
${}^{240}Am$			α 2n	< 200 μ b
$15_N + {}^{235}U$	80 MeV	${}^{246}Es$	4n	< 1.0 μ b
		${}^{246}Cf$	p3n; ${}^{246}Es \xrightarrow{EC}$	1.3 μ b

C. Spontaneous Fission Experiments and Results

The bombardments performed to search for spontaneous fission activity in even-even neutron deficient actinide isotopes are listed in Table 4. No spontaneous fission activities were seen with half-life limits less than approximately 15 hours and greater than several hundred nanoseconds in any of the bombardments. Using the method described previously, crude estimates were made for spontaneous fission branching ratios and spontaneous fission half-lives, shown also in Table 4. The assumed cross-sections were taken to be about 15 times lower than those calculated by JORPLE to try to give a more realistic picture of the experimental cross-section for neutron deficient actinides. This factor was arrived at by comparing a number of different experimental cross-sections in this actinide mass region with calculations.

Table 4.

Summary of Spontaneous Fission Bombardments

Reaction	Isotope	σ_{calc}^*	%SF _{EXPT}	$T_{1/2}^{\dagger}$ _{SF-EXPT}	$T_{1/2}^{\dagger}$ _{SF-PRED}
$^3\text{He} + ^{233}\text{U}$	^{228}Pu	0.1nb	< 90%	> 2m	1.5y
$^{24}\text{Mg} + \text{NAT'L Pb}$	^{228}Pu	735nb	< 9%	> 22m	
$^3\text{He} + ^{233}\text{U}$	^{230}Pu	9nb	< 5%	> 4hr	$2.1 \times 10^3\text{y}$
$^{28}\text{Si} + ^{208}\text{Pb}$	^{232}Cm	15nb	< 30%	> 3.3m	28m
$^{40}\text{Ar} + \text{NAT'L Hg}$	^{238}Cf	1.3nb	< 25%	> 4sec	260ns

* Calc. from JORPLE with corrections as described in text.

† Limits are based on the assumed cross-section and half-life.

+ Values obtained from Figure 3.

VII. DISCUSSION

A. Berkelium-242

From the data presented there seems little reason to doubt that the observed 7-minute component of the curium X-rays found in the berkelium chemical fraction is due to ^{242}Bk . The most convincing evidence is the presence of ^{242}Cm in the berkelium chemical fraction. The only way it could be present is from the decay of the ^{242}Bk parent. The shape of the excitation functions, the projectile energy for σ_{max} , the decay mode, the half-life value, and the cross-bombardments all lend additional support to the conclusion that this activity is indeed ^{242}Bk .

An important question is: Is this the ground state of ^{242}Bk or is it an isomeric state? Certainly an explanation of this activity as an isomer of berkelium would help explain the low cross-section (for this case). However, the experimental cross-sections for a number of neutron deficient isotopes are also low compared with JORPLE calculations as shown in Table 5. Thus the low experimental cross-section does not appear to be out of line with other reported results. In order to examine the question of whether or not this species is an isomeric state, X-ray/gamma coincidence should be performed. This would reveal any correlations between gamma emission and the electron capture decay of the isotope, i.e. whether or not the decay was to the ground state of curium. A major obstacle to this attempt is the low production cross-section of the isotope. It is unlikely that enough activity can be produced to yield any useful information, especially if chemistry must be performed. Perhaps the best possibility would exist with an on-line mass separator

which could separate the berkelium and transport it to a coincidence detection system in a minimum amount of time. The limiting factor here would be the degree of separation and transportation efficiency.

Table 5.

Reaction	$\sigma_{\text{EXP'L}}$	$\sigma_{\text{JORPLE}}^{\dagger}$	Ref.
$^{232}\text{Th}(^{11}\text{B}, 5n)^{238}\text{Am}$	120 μb	2000 μb	133
$^{232}\text{Th}(^{12}\text{C}, 4n)^{240}\text{Cm}$	40 μb	185 μb	134, 135
$^{238}\text{U}(^{10}\text{B}, 4-5n)^{244, 243}\text{Bk}$	14 μb	414 μb	*
$^{238}\text{U}(^{11}\text{B}, 4n)^{245}\text{Bk}$	< 20 μb	1200 μb	*
$^{235}\text{U}(^{11}\text{B}, 4n)^{242}\text{Bk}$	10 μb	200 μb	*
$^{233}\text{U}(^{12}\text{C}, 5n)^{240}\text{Cf}$	$\sim 0.007\mu\text{b}$	0.150 μb	36
$^{233}\text{U}(^{12}\text{C}, 4n)^{241}\text{Cf}$	$\sim 0.040\mu\text{b}$	1.4 μb	36
$^{234}\text{U}(^{12}\text{C}, 5n)^{241}\text{Cf}$	$\sim 0.015\mu\text{b}$	0.740 μb	36

[†] It must be stressed that these disagreements are probably local perturbations and that, in general, JORPLE provides quite useful information about actinide cross-sections.

* This work.

Another explanation for the low cross-section could lie in entrance channel effects, i.e. projectile break-up due to the Coulomb field of the target nuclei. While from this work there is no evidence for such a problem, it is well-known that both ^6Li and ^7Li are predisposed to break-up into ^3He , p2 n and α , p2 n, respectively.^{136, 137} Unternährer¹³⁸ and co-workers have demonstrated ^9Be break-up, and Ollerhead¹³⁷ reported limited data on the break-up of $^{10, 11}\text{B}$. If dissociation of the projectile were to occur prior to the formation of the compound nucleus, the

fusion yields would certainly be lower than expected. In fact, it has been found, both in this work and by others,³⁵ that fusion yields from boron reactions are uniformly lower than expected.

An additional contribution might also come from the fissionability of the target. Admittedly, ^{232}Th and ^{238}U are not considered very fissionable; however, ^{233}U and ^{235}U are. The compound nucleus yield would certainly be reduced if the target nuclei were to undergo fission prior to or during fusion. This explanation seems unlikely, however, since the reactions $^{234}\text{U}(^{12}\text{C},4n)^{242}\text{Cf}$; $^{235}\text{U}(^{12}\text{C},5n)^{242}\text{Cf}$; and $^{235}\text{U}(^{12}\text{C},4n)^{243}\text{Cf}$ all have cross-sections which agree quite well with JORPLE calculations.

Assuming that the above mentioned contributions are small, the most likely explanation for the cross-section discrepancies lies in the calculations themselves. As has been mentioned, the values used for such parameters as d , r_0 , etc. were determined from best fits to the data available (1973) for the whole actinide region. It is quite possible that these parameters are not applicable in the neutron deficient region. The most likely parameter to be in error would be Γ_n/Γ_f . However, the change required if this is the only value in error is probably too great (lower by approximately a factor of 3) and yields a somewhat unrealistic Γ_n/Γ_f ratio.^{8,139,140} More probably, small changes in a number of parameters are required rather than a drastic change in any one.

B. (H,1.,pxn) and (H,1.,axn) Reaction Products

In 1967 Fleury et al.^{141,142} measured the production of ^{240}Cm and ^{242}Cm from $^{10,11}\text{B}$ on ^{238}U . They interpreted the excitation functions

to be due to a compound nucleus mechanism. Presumably, ^{242}Bk and ^{240}Bk were produced which then decayed to their curium daughters prior to the chemical separations. The cross-sections they reported were significantly higher than those measured in this work for ^{242}Bk . In light of the newly measured cross-sections for ^{242}Bk , it did not seem reasonable that cross-sections as high as several hundred microbarns for ^{242}Cm could have come only from the decay of ^{242}Bk as assumed by Fleury. Indeed, this work has shown that such is not the case. Only a small contribution to the curium cross-section comes from the decay of berkelium. The primary yield of ^{242}Cm (and presumably ^{240}Cm) has been shown to be due to either direct production of ^{242}Cm (^{240}Cm) or to the evaporation of a proton and 3 (5) neutrons from the compound nucleus. The latter seems extremely unlikely due to the high Coulomb barrier to charged particle emission from a nucleus with a Z as high as 97.

This work further demonstrates the high yields of axn products in heavy ion reactions in the actinide region. The reaction showing the highest yield is the $\alpha 2n$ followed by the $\alpha 3n$. Again, it is unlikely that these products result from alpha evaporation, but rather, they are most probably the result of direct formation. To this effect, Hahn et al.¹⁷ have demonstrated that axn products from $^{12}\text{C} + ^{239}\text{Pu}$ are indeed the result of a transfer mechanism rather than a compound nucleus mechanism. They found, as well, that the $\alpha 2,3n$ product yields (Cf) were much higher than the 2-4n compound nucleus yields (Fm). Actually this is not too surprising since Γ_n/Γ_f is so highly Z dependent. The (H.I.,pxn) and (H.I.,axn) products have Z's lower than the xn products and, therefore, higher Γ_n/Γ_f ratios. While more compound nuclei may be formed initially,

fewer of them are able to survive fission competition due to their lower Γ_n/Γ_f ratios. Of particular note here is that the yields of the $\alpha, 2n$ products from $^{12}\text{C} + ^{239}\text{Pu}$ are within a factor of 2-3 of those reported for the compound nucleus reaction from $^{12}\text{C} + ^{238}\text{U}$ ¹⁴³ producing the identical californium isotopes! Kutznetsov et al. ^{144, 145} also have reported αxn reactions with cross-sections four times larger than the corresponding compound nucleus reaction using boron as the projectile; Bimbot et al. ¹⁴ find significant αxn yields with ^{12}C projectiles; and Brandshtetr et al. ¹⁴⁶ find the $\alpha 4n$ (^{246}Cf) product from $^{16}\text{O} + ^{238}\text{U}$ to be equal in yield to the $4n$ (^{246}Cf) product from $^{12}\text{C} + ^{238}\text{U}$.

The same trends were found in this work using boron and nitrogen projectiles on uranium and thorium targets. The compound nucleus cross-sections are significantly lower than pxn and αxn products from the same reaction. While no data were measured to confirm that these products result from direct reactions, it seems reasonable that such is the case. The particle "transferred" to the ^{235}U target in the boron bombardments to produce americium would be ^7Li followed by neutron emission. Hubert et al. ¹⁴⁷ found very high yields of lithium transfer products in studies of boron on tantalum. ^8Be would be the particle transferred to form curium isotopes. As mentioned, Hahn ¹⁷ has studied ^8Be transfers and found their yields to be significant.

These results suggest that, in some cases at least, it may be more profitable to attempt to produce a neutron deficient actinide isotope via a transfer mechanism (p or αxn) than by a compound nucleus mechanism. Such is the method used by Kutznetsov. ^{144, 145} Of perhaps even greater

importance is the effect these data have on the " $Z_1 + Z_2 = \text{New Element}$ " hypothesis of Flerov and co-workers¹⁴⁸⁻¹⁵⁴ at Dubna. These researchers rely heavily upon spontaneous fission data for identification of new isotopes and elements which, as mentioned before, does not provide unambiguous proof of either Z or A for a spontaneous fission activity. It has often been claimed by this group in defense of their identification of various new elements and isotopes, that (H.I.,pxn) and (H.I., α xn) reactions do not occur to any great degree in regions of high Z, and that, therefore, it is reasonable to assume the resulting spontaneous fission activity is due to the compound nucleus, i.e., $Z_1 + Z_2 = Z_1 + 2$. While it may be true that proton or alpha evaporation does not occur to any degree, direct transfer reactions giving the same products as proton or alpha evaporation reactions do occur and to a degree often in excess of the compound nucleus! Nor are excitation functions or angular distribution measurements necessarily sufficient for elimination of a transfer mechanism. For example, Fleury's^{141,142} excitation function for $^{238}\text{U}(^{10}\text{B},?)^{242}\text{Cm}$ looks deceptively like a compound nucleus excitation function, and Hahn et al.¹⁷ have cautioned that at certain bombarding energies it may not be possible to distinguish the angular distributions of compound nucleus from transfer reactions.

C. Spontaneous Fission Studies

In the spontaneous fission experiments the only point of interest is that of californium. The points for ^{228}Pu , ^{230}Pu , and ^{232}Cm lie below the predicted half-lives so no information was gained about their relation to these curves (Figure 3). The number of beam particles

necessary to raise the limits determined for plutonium and curium would require more beam time than is currently available for this type of experiment.

The lower limit to the spontaneous fission half-life for ^{238}Cf has been determined to be several seconds, while the extrapolated parabola yields a prediction of several hundred nanoseconds (Figure 3). The shortest electron capture half-life predicted for this isotope is 0.6 sec.⁴² Therefore, if the spontaneous fission half-life were to follow these predictions and be several hundred nanoseconds, then the primary decay mode seen would be spontaneous fission. Since such is not the case, the spontaneous fission half-life is either not as short as predicted, or much, much shorter than predicted; or the alpha/electron capture half-life is much shorter than predicted, i.e. much shorter than the spontaneous fission half-life.

Undoubtedly, the straight extrapolation assumption is much too naive. Indeed, theoretical calculations of Viola and Wilkins¹⁵⁵ predict the spontaneous fission half-life for ^{238}Cf to be on the order of days! Their predictions are derived from experimental data using the liquid drop model, modified to account for nuclear deformation and level structure effects. Even-even isotopes were taken as reference nuclei. Their calculations show that fission lifetimes and barriers decrease regularly with increasing values of Z^2/A , as predicted by the liquid drop model, but they do not decrease as dramatically as the empirical extrapolations in Figure 3 would predict. Deviations from their predictions are the result of irregularities in nuclear structure caused by single particle effects, deformation, pairing interactions, etc.

It is important to realize that these results do not eliminate the possibility of the existence of spontaneous fission isomers of the isotopes studied, which typically have half-lives shorter than several milliseconds, but probably only reflect the non-existence of spontaneous fission as the primary decay mode from the ground state within the limits determined.

While no conclusions can be reached regarding the true spontaneous fission half-lives of the isotopes examined, it seems clear that a simple extrapolation of the known points is not sufficient to predict spontaneous fission half-lives, a point often argued by the groups involved with the $^{260}_{104}$ identification and half-life controversy.^{150,156,157}

VIII. CONCLUSIONS

Evidence has been presented for the new berkelium isotope, ^{242}Bk . Originally it was hoped that ^{241}Bk and perhaps even ^{240}Bk would also be found. Most probably, the combination of a short half-life and a small production cross-section prevented the observation in these experiments of the lighter isotopes. Future experiments should focus on the shortest possible chemical separation--perhaps even on line--coupled with as sensitive an X-ray detection system as possible. While the cross-section for ^{242}Bk ($\sim 10 \mu\text{b}$) was small, it still should be possible to observe ^{241}Bk with the proper sensitivity.

A major problem would exist, however, if ^{241}Bk decays by electron capture with a half-life very close to that of ^{242}Bk . In that case, it may not be possible to resolve this isotope from ^{242}Bk if its cross-section is less than several microbarns. In this situation, some sort of mass separation system, as mentioned earlier, would be appropriate. As before, the limiting factors would be cross-section and transportation/separation efficiency.

Of particular importance is the result that yields of (Heavy Ion, pxn) and (Heavy Ion, axn) transfer products are equal to or exceed the yields of the compound nucleus products in the actinide region. As has been emphasized, these findings point out the dangers in assuming yields of pxn and axn products to be negligible in the heavy element region. While charged particle evaporation may be minimal in this Z region, one cannot neglect the production of the same products by a direct mechanism. Before a new element or isotope may be claimed, it must be

shown that such directly produced products are not responsible for the new observations.

A systematic study of (H.I.,pxn) and (H.I.,αxn) versus (H.I.,xn) reactions should be performed to further examine this phenomenon. Projectiles and targets should be varied to look for mass effects, even-odd effects, Z effects. Heavier projectiles should be used to examine the difference between them and lighter projectiles.

As the Z of the products increases, the experiments become increasingly more difficult. The cross-sections are dramatically lower for the production of, for example, fermium compared to curium. Further, the half-lives of the most interesting products are on the order of minutes rather than hours or days making chemical separations, especially among the actinide elements, very difficult. Examples of systems that might provide useful information would be:

Once a systematic study of the actinides has been completed, the study could be expanded to include the region around lead where fission is no longer eliminating most of the products.

(#) Examined in part by Hahn et al.¹⁷

While the lack of observations in the spontaneous fission experiments was at first disappointing, it was not too surprising. The sensitivity for three of the isotopes was insufficient to draw any conclusions, while the results for ^{238}Cf do not rule out a very, very short half-life ($< \sim 100$ ns). What is of importance, however, is the fact that the half-lives do not appear to follow strictly empirical predictions on the neutron deficient side of the 152-neutron subshell. Future experiments should concentrate on the higher Z elements since these have the greatest possibility of being observed, i.e., require less beam intensity for the experiments. The drum speed should be increased as much as possible to search for extremely short half-lives. Decay-in-flight techniques could be used to reach even shorter half-lives. Unfortunately, however, if a short-lived activity is found, there is no simple way using the present methods to distinguish it from an isomer of the same or even another element. As has been mentioned time and again, spontaneous fission measurements alone are inconclusive regarding the Z and mass of an activity.

In conclusion then, this work has pointed out the grave errors in assuming the identity of a new element or isotope based solely on the assumption that $Z_1 + Z_2 = Z_{1+2}$, especially when spontaneous fission is the only method of identification. It further cautions those scientists who doggedly insist on sticking to empirical systematics that such predictions, while often quite useful, are not infallible and are at times subject to question.

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XBL 786-1080

Fig. 1

CBR 783-3608

Fig. 2

XBL766-8233

Fig. 3

XBL766-8234

Fig. 4

XI 779-1817A

Fig. 5

Fig. 6

XBL 786-9224

XBL 778-1633A

Fig. 7

FIG. 8

XBL 786-1079

TTA SOLVENT EXTRACTION OF BK

XBL 786-1076

Fig. 9

HDEHP SOLVENT EXTRACTION OF Bk

XBL 786-1075

Fig. 10

TRANSURANIUM SEPARATION

XBL 786-1077

Fig. 11

XBB 694-2366

Fig. 12

COUNTS PER CHANNEL

8K-111

Fig. 14

XRL 786-9230

XBL 786-9229

Fig. 15

Fig. 16

XBL 786-1078

XBL 786-9223

Fig. 17

Fig. 18

XBL 786-9221