New isotope ^{242}Bk

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A new isotope of berkelium has been identified through bombardments of ^{235}U with ^{11}B, ^{238}U with ^{10}B, and ^{232}Th with both ^{14}N and ^{15}N. The new activity decays by electron capture with a half-life of 7.0 ± 1.3 minutes. The α-branching ratio for this isotope is less than 1% and that for spontaneous fission is less than 0.03%. Analysis of the excitation functions, the chemical behavior of the isotope, and the milking of the ^{242}Cm daughter leads to the positive identification of this isotope as ^{242}Bk. A preliminary search was made for ^{241}Bk but was unsuccessful. Examination of the (heavy ion, *pxn*) (Cm) and (heavy ion, αxn) (Am) transfer products in comparison with the (heavy ion, *xn*) (Bk) compound nucleus products produced in the above reactions revealed transfer reaction cross sections to be equal to or greater than the compound nucleus cross sections. The data suggest that in some cases the yield of an isotope produced via a *pxn* or αxn transfer reaction might be higher than its production via an *xn* compound nucleus reaction.

[NUCLEAR REACTIONS Compound nucleus, proton and alpha transfer reactions: ^{235}U(^{10,11}B, *X*)*I*; ^{238}U(^{10}B, *Q*)*R*; ^{232}Th(^{14,15}N, *M*)*T*; measured excitation functions for ^{242}Bk; measured σ for ^{238}Am, ^{239}Am, ^{240}Am, ^{240}Cm, ^{242}Cm, ^{243}Bk.]

I. INTRODUCTION

Recently, extensive efforts have concentrated on promising mechanisms for the production of superheavy elements.[1,2] Very little work, however, has concentrated on investigating neutron deficient actinide isotopes. Berkelium, in particular, has been ignored. Until this work, the lightest isotope of berkelium known was ^{243}Bk, the first berkelium isotope discovered,[3] although there is some evidence for a spontaneously fissioning ^{242}Bk isomer.[4]

In this paper we report on our investigation of the electron capture decay of neutron deficient berkelium isotopes produced in reactions of ^{10}B and ^{11}B with ^{235}U, ^{10}B with ^{238}U, and ^{14}N and ^{15}N with ^{232}Th. We report also on the high yields of (H. I., pxn) and (H. I., αxn) transfer products observed in comparison to the yields of (H. I., *xn*) compound nucleus products.

II. EXPERIMENTAL

The targets were prepared by electrospraying[5] the nitrates dissolved in acetone onto 1-mil thick beryllium backing foils kept at ~250 °C. The 0.5-cm^2 area targets were heated to red heat in an induction furnace to ensure conversion of the target material to the oxides. The uranium targets were isotopically enriched—^{238}U, 99.9% (*Q* material), ^{235}U, 94.1%—while the thorium isotopes were present in natural abundance. The typical target thickness was ~400 μg/cm^2.

The ^{10}B^{+3}, ^{11}B^{+3}, ^{14}N^{+4}, and ^{15}N^{+4} beams of energies from 63 to 112 MeV provided by the 88-inch cyclotron at the Lawrence Berkeley Laboratory were collimated to a 0.4-cm^2 area and degraded, before entering the target, to energies between 52 and 93 MeV (Fig. 1). The degrading foils were beryllium or havar and served also as the upstream window of the target cooling system. The beam intensities were between 2.5 and 5 μA.

Target cooling was accomplished by passing cold nitrogen gas from liquid nitrogen between the target and an upstream window of havar or beryllium. The berkelium products recoiling from the target were caught in a thin aluminum catcher foil which was cooled with 150 Torr of helium and was located 0.5 cm from the target. After passing through the target assembly and the catcher foil, the beam was stopped in a Faraday cup. Two rare-earth magnets were placed in front of the target to avoid inaccurate beam readings due to secondary electrons.

Immediately following the irradiation, the catch-

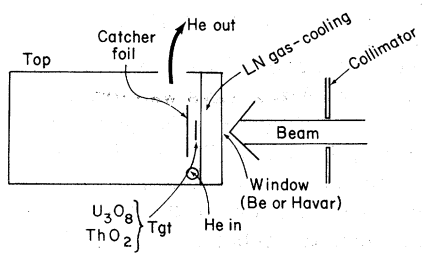

FIG. 1. Schematic of the Recoil Collection Irradiation System.

er foil was dissolved in hot 8M HNO_3 in the presence of Hg^{+2}. The berkelium products were oxidized to +4 using CrO_3 and were extracted into bis (2-ethylhexyl) orthophosphoric acid (HDEHP) in n-dodecane.[6] After being back-extracted into 1.5M H_2O_2/10M HNO_3, the berkelium products were plated on 1-inch diameter platinum disks for x-ray, γ, and/or α counting. The time from the end of the bombardment (eob) to the start of counting was 12–14 min with a chemical yield of ~50%.

Curium/americium samples were extracted using a standard transplutonium chemistry consisting of: lanthanum carrier and fluoride precipitations, a Dowex 1 × 8 HNO_3 column, a saturated HCl MP-50 column, a $BaCl_2$ precipitation, a Dowex 1 × 8 HCl column, and a thenoyltrifluoroacetone (TTA) extraction of the final curium/americium fraction.[7,8]

α particles were measured with 300-mm^2 gold-plated surface barrier detectors which were calibrated using a combination of thin ^{233}U (4.824 MeV), ^{241}Am (5.485 MeV), ^{244}Cm (5.805 MeV), and ^{252}Cf (6.118 MeV) sources and a linear pulse generator. After suitable amplification, the pulses were fed into a 4096-channel pulse-height analyzer which was divided into 1024-channel sections allowing simultaneous use of four separate detectors. The absolute detection efficiency for ^{241}Am was typically 20% and the full width at half maximum (FWHM) at 5.485 MeV was 20 keV.

x and γ rays were analyzed using a shielded 60-cm^3 volume coaxial-germanium, lithium drifted, diode detector connected, after amplification, to a 4096-channel pulse-height analysis system. Energy calibrations and detector efficiencies were measured using an NBS Mixed Radionuclide Gamma Ray Emission-Rate Point Source Standard (SRM 4216-B) which contained nine nuclides covering energies from 88.03 keV (^{109}Cd) to 1332.48 keV (^{60}Co). The resolution for the curium $K\alpha_1$ x-ray peak (109.27 keV) was 1.5 keV FWHM and the efficiency for this peak when measured 0.5 cm from the detector face was 15%. A sample x-ray spectrum is shown in Fig. 2.

III. RESULTS

Evidence for a short-lived component in the curium $K\alpha_1$ and $K\alpha_2$ x-rays was found in the bombardment of ^{235}U with ^{11}B at 60 MeV lab even though the time from the end of the bombardment to the start of counting was 25 min. Subsequent bombardments using a shortened chemistry confirmed this component as having a half-life of 7.0 ± 1.3 min. A typical decay curve of the curium $K\alpha_1$ and $K\alpha_2$ x rays is shown in Fig. 3. The short-lived component stands out quite strongly and is easily resolved from the longer-lived activity of ^{243}Bk

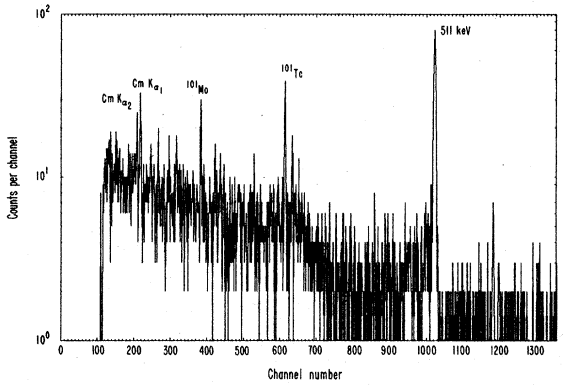

FIG. 2. A typical x-ray spectrum of a berkelium sample from ^{11}B + ^{235}U.

($t_{1/2}$ = 4.5 h). The excitation function for this activity from ^{11}B on ^{235}U, shown in Fig. 4, agrees quite well in peak energy and width with calculations of Alonso[9] for the (^{11}B, 4n) reaction. However, the experimental cross section, σ_{max} = 10 ± 2 μb, is approximately a factor of 20 below that calculated.

As a further check on the mass number and on the measured cross section for this short-lived activity, ^{232}Th was bombarded with ^{15}N. Once again, the experimental excitation function, Fig. 5, agrees well in energy and width with calculations, but the σ_{max} = 9 ± 1 μb is ~25 times lower than predicted.

The 7-min activity was also produced in the reactions ^{10}B on ^{238}U (σ_{max} = 8.9 μb) and ^{14}N on ^{232}Th (σ_{max} = 0.48 μb). The bombarding energy in the ^{10}B case corresponded to a 6n reaction and in the ^{14}N case to a 4n reaction. Once more the cross sections were lower than expected by approxi-

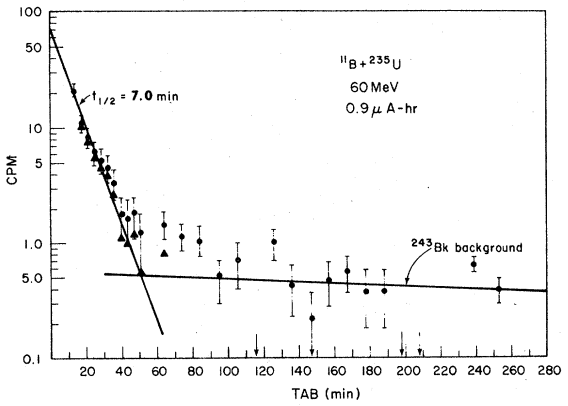

FIG. 3. Decay of curium $K\alpha_1 + K\alpha_2$ x rays from the berkelium chemical fraction (^{11}B + ^{235}U).

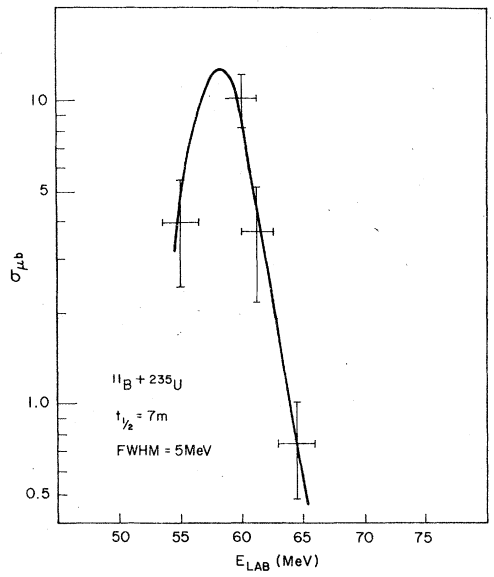

FIG. 4. Excitation functions for ^{242}Bk from ^{11}B + ^{235}U. The solid curve represents the Alonso calculations lowered in cross section approximately by a factor of 20.

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A search was made for ^{241}Bk by bombarding ^{235}U with ^{10}B at energies from 52–60 MeV. This energy range should have maximized ^{241}Bk production while the yield of ^{242}Bk should have been several orders of magnitude lower. No activity with a half-life greater than 3 min and a cross section greater than 2 μb was observed.

One of the best ways to confirm the Z and A of a new isotope is to identify positively its daughter

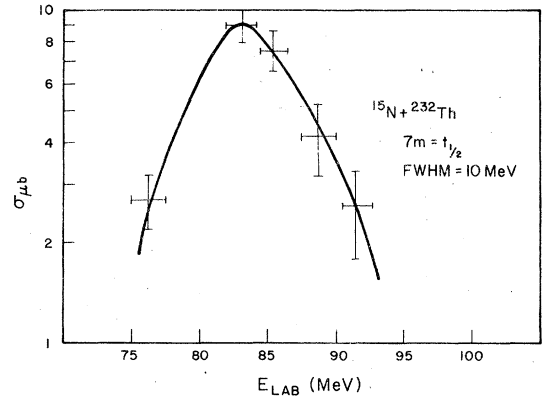

FIG. 5. Excitation function for ^{242}Bk from ^{15}N+^{232}Th. The solid curve represents the Alonso calculations lowered in cross section approximately by a factor of 25.

in the original parent chemical fraction. Berkelium chemical fractions from a number of separate bombardments were combined and a curium fraction was chemically separated from this pool to look for the ^{242}Cm daughter of ^{242}Bk. Twelve 25-min bombardments were performed using ^{11}B on ^{235}U at 60 MeV lab. Berkelium was separated from the dissolved catcher foil within 4.5 min from the end of the bombardment and the final berkelium fractions from all the runs were pooled. A ^{244}Cm tracer was added and a day later a curium fraction was removed from this combined berkelium pool with a chemical yield of 70%. From the amount of ^{242}Cm present (Fig. 6) the cross section for ^{242}Bk was found to be 9.3 ± 1.5 μb, in excellent agreement with the direct measurements. The ^{242}Cm was identified on the basis of both α energy and half-life. The ^{241}Am tracer was used to calibrate the columns.

It was conceivable that the lower than expected cross sections could have been due to the decay of ^{242}Bk by α particle emission or even spontaneous fission. A number of berkelium fractions were α counted within 8 min from the end of the bombardment. No α particle activities at all were observed in the first few hours of counting. Several catcher foils were counted 1 min after the end of the bombardment. As in the chemistry runs, no (unidentified) α particle activities were seen. Direct γ- and x-ray counting of the catcher foils was attempted but was unsuccessful due to the intense β-, γ-, and x-ray background in the foils.

That the light berkelium isotopes do not decay by spontaneous fission has shown by Somerville.[10] He bombarded uranium isotopes with ^{10}B and ^{11}B at various energies and no unidentified fission activity was observed resulting in the deduction of a fission branch of less than 0.03% for ^{242}Bk. Eastham and Grant[11] bombarded ^{232}Th with ^{14}N at the energy corresponding to the peak yield for the

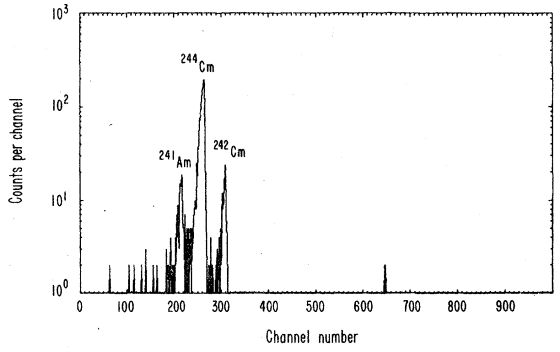

FIG. 6. α spectrum of the ^{242}Cm milking sample from ^{11}B+^{235}U at 60 MeV (10-day count).

$4n$ reaction and set a cross-section upper limit for the spontaneous fission of ^{242}Bk at 50 nb for a half-life greater than 2 ns.

Since the cross sections measured for the (H.I., xn) compound nucleus reactions were much lower than expected, the question was raised as to what products were being produced. It was decided to examine the (H.I., pxn) and (H.I., αxn) products, curium and americium respectively, produced directly in the same reactions and at the same energies used to produce ^{242}Bk. In the first experiment ^{235}U was bombarded with a total of 16.9 μA h of $^{11}B^{+3}$. Curium and americium were chemically separated from the catcher foil and the final chemical fraction was analyzed for the emission of both γ rays and α particles.

The results are given in Table I. ^{238}Am, ^{239}Am, and ^{240}Am were detected by γ-ray analysis, while ^{240}Cm and ^{242}Cm were detected by emission of α particles. The berkelium data were not determined in the same bombardments 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 the (H.I., xn) compound nucleus mechanism. The total contribution of ^{242}Am decay to the production of ^{242}Cm should be very small since the emission or transfer of only one α particle and no neutrons or other particles has a very small probability, especially at excitation energies greater than 35–40 MeV. Data were also obtained from reactions of ^{10}B with ^{238}U and ^{14}N with ^{232}Th, as shown in Table I. In the ^{10}B case only information about products resulting from the loss of a proton and a few neutrons was gained since the products resulting from the loss of an α particle and a few neutrons were too long-lived to be seen.

IV. DISCUSSION

A. Berkelium-242

From the data presented, there seems little reason to doubt that the observed 7-min 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 proper amount in the berkelium chemical fraction. The only way it could be present is from the decay of the ^{242}Bk parent. Both the decay mode and half-life for this new activity are in good agreement with predictions[12,13] for ^{242}Bk. Thus, the shape of the excitation functions, the projectile energy corresponding to σ_{max}, the decay mode, the value of the half-life, and the cross bombardments all lend additional support to the conclusion that this activity is indeed ^{242}Bk.

A bit of concern was raised over the discrepancy between the calculated cross sections and those measured experimentally. The cross section calculation code employed, JORPLE, developed by Alonso *et al.*[9,14] and Rasmussen[15] and based on a computer code of Sikkeland and Lebeck,[16] 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 Thomas[17] and of Rasmussen *et al.*,[14,15] to angular momentum effects, to nuclear deformation, to fusion probability using the Hill-Wheeler transmission formula,[18] to compound nucleus formation using the formula of Blatt and Weisskopf,[19] and finally to deexcitation of the compound nucleus by neutron evaporation which is based on the Jackson model,[20] corrected for fission competition using the empirical formula of Sikkeland *et al.*[21] for Γ_n/Γ_f, the ratio of neutron emission to fission.

As in all calculations of this sort, the cross sections obtained are extremely 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 available 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 the 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 excitaton functions calculated from this code agree quite well in shape, energy, and cross section with most of the experimental data in the actinide region. Unfortunately, however, there appear to be large discrepancies in cross sections in the region of neutron deficient actinide isotopes. Williams[22] lists a number of cases where the experimental cross sections are factors of 20–30 below those calculated. Thus, it does not appear that the low cross sections measured in this work are out of line with other reported results. As mentioned, the parameters used in these calcula-

TABLE I. Yield of pxn and αxn products.

System	Energy	Product	Reaction	Cross section
$^{11}B+^{235}U$	60 MeV	^{242}Bk	$4n$	10 μb
		^{243}Bk	$3n$	1
		^{242}Cm	$p3n$	
			$4n;^{242}Bk \xrightarrow{ec}$	78
			$\alpha;^{242}Am \xrightarrow{\beta^-}$	
		^{242}Cm	$p3n$	
			$\alpha;^{242}Am \xrightarrow{\beta^-}$	68
		^{240}Cm	$p5n$	0.035
		^{240}Am	$\alpha 2n$	1500
		^{239}Am	$\alpha 3n$	410
		^{238}Am	$\alpha 4n$	1.6
$^{10}B+^{238}U$	67 MeV	^{242}Bk	$6n$	8.9
		^{242}Cm	$p5n$	
			$6n;^{242}Bk \xrightarrow{ec}$	60
			$\alpha;^{242}Am \xrightarrow{\beta^-}$	
		^{242}Cm	$p5n$	
			$\alpha;^{242}Am \xrightarrow{\beta^-}$	51
		^{240}Cm	$p7n$	0.12
$^{14}N+^{232}Th$	77 MeV	^{242}Bk	$4n$	0.48
		^{242}Cm	$p3n$	
			$4n;^{242}Bk \xrightarrow{ec}$	1.00
			$\alpha;^{242}Am \xrightarrow{\beta^-}$	
		^{242}Cm	$p3n$	
			$\alpha;^{242}Am \xrightarrow{\beta^-}$	0.52
		^{240}Cm	$p5n$	0.055
		^{240}Am	$\alpha 2n$	200

tions were obtained from best fits to a wide range of actinide data. It is quite possible that one or more of the values used is incorrect for this neutron deficient region of the actinides. The most likely parameter value to be in error is Γ_n/Γ_f. This value is determined from an empirical formula of Sikkeland[21] which is based only on data available though 1967. The change required in Γ_n/Γ_f to match the experimental cross section is to lower the empirical value by approximately a factor of 3. Such a change falls within the errors allowed in Γ_n/Γ_f by Gavron *et al.*[23] for the neutron deficient actinide isotopes.

B. (H.I., pxn) and (H.I., αxn) reaction products

In 1967 Fleury *et al.*[25,26] measured the production of ^{240}Cm and ^{242}Cm produced in the reactions of ^{10}B and ^{11}B with ^{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 our measurements, it did not seem reasonable to us that cross sections as high as several hundred μb 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 operating against charged particle emission from a nucleus with a Z as high as 97.

This work further demonstrates the high yields of products resulting from the loss of an α particle from the compound nucleus in heavy ion reactions in the actinide region. The reaction showing the highest yield is the $\alpha 2n$ reaction while the next highest yield is found with the $\alpha 3n$ reaction. Again, it is unlikely that these products result from α particle evaporation, but rather, they are most probably formed via a transfer mechanism. To this end, Hahn *et al.*[27] have demonstrated that αxn products from $^{12}C+^{239}Pu$ 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 $2,3,4n$ compound nucleus yields (Fm). This is not too surprising since Γ_n/Γ_f is lower for fermium than for californium leading to an increase in the number of fermium nuclei lost to fission.

Of particular note here is that the yields of the

$\alpha 2$, $3n$ products from $^{12}C + ^{239}Pu$ are within a factor of 2–3 of those reported for the compound nucleus reaction from $^{12}C + ^{238}U$ producing the identical californium isotopes.[28] Bimbot *et al.*[29] also find significant αxn yields from ^{12}C projectiles. Further, Brandshtetr *et al.*[30] have found the $\alpha 4n$ (^{246}Cf) product from $^{16}O + ^{238}U$ to be equal in yield to the $4n$ (^{246}Cf) product from $^{12}C + ^{238}U$. Perhaps most interesting is that Kutznetsov *et al.*[31,32] have reported αxn reaction products with cross sections four times larger than the corresponding compound nucleus reaction using boron as the projectile.

These 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 those for pxn and αxn products from the same projectile-target combination. While no data were measured to confirm that these products result from direct or transfer 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 ^{7}Li followed by neutron emission. In support of this, Hubert *et al.*[33] found very high yields of lithium transfer products in studies of boron on tantalum. ^{8}Be would be the particle transferred to form curium isotopes from a ^{235}U target. Hahn[24] has studied ^{8}Be transfers in this actinide region and found their yields to be significant.

V. SUMMARY AND COMMENT

In summary, then, a new isotope of berkelium, mass number 242, has been produced in reactions of boron on uranium and nitrogen on thorium with a $\sigma_{max} = 10$ μb. It decays by electron capture with a half-life of 7.0 ± 1.3 min. Yields of (H.I., pxn) and (H.I., αxn) transfer products were found in excess of the compound nucleus products suggesting that in some instances it may be more profitable to produce neutron deficient actinide isotopes via a (H.I., p or αxn) transfer mechanism rather than via a (H.I., xn) compound nucleus mechanism.

Perhaps a last comment should be added concerning the effect these results may have on the current problems connected with claiming the discovery of new elements and/or isotopes. There are currently two major modes of identification for new elements which are too short-lived to be chemically separated. One method employs spontaneous fission detection techniques, while the other method relies on α particle identification and, where possible, genetic links to known daughters and granddaughters. The use of spontaneous fission measurements as the sole identification method for new elements suffers from a severe drawback: No positive A *or* Z identification of the decaying species is provided. This is in contrast to α particle analysis where genetic links can be observed during the decay of the parent and daughters leading to both positive Z and A identification of the newly observed activity. Since the spontaneous fission technique does not give unambiguous information concerning Z or A, the assumption is often made that the new activity observed is the result of a compound nucleus mechanism which has long been considered to produce activities in the highest yield, i.e., the assumption is made that $Z_1 + Z_2 = Z_{1+2}$. The yield of proton or α *evaporation* products is reasonably assumed to be negligible in this high Z region. Therefore, when a new spontaneous fission activity is produced in a heavy ion reaction, the claim is often made that a new element or isotope has been produced. However, data from this study and from the other work referenced here show that while the assumption that proton and α particle *evaporation* product yields are negligible may be valid, the assumption that the yields of these very same products produced via a transfer mechanism are negligible is not. In fact, it now has been shown that the (H.I., pxn) and (H.I., αxn) product yields can be as much as 100 times greater than the (H.I., xn) product yields from the same projectile-target combination. It is not valid to assume that the activities produced in the greatest yields are due to a compound nucleus mechanism in heavy ion reactions. Therefore, the observation of a new spontaneous fission activity in a heavy ion bombardment *may*, indeed be due to the compound nucleus, but it may have an even greater probability of being due to an element one or two Z units lower than the compound nucleus. These results emphasize the great care that must be taken when a new activity is to be identified. With only spontaneous fission as the method of detection, it is not possible to determine uniquely the Z or the A of the new activity. Before a new element can be claimed, it must be shown that directly produced nuclei, i.e., noncompound nuclei, are not responsible for the new observations, especially when doubt may exist about the Z identification of the new discovery.

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