Beta-delayed fission and neutron emission calculations for the actinide cosmochronometers

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The Gamow-Teller beta-strength distributions for 19 neutron-rich nuclei, including ten of interest for the production of the actinide cosmochronometers, are computed microscopically with a code that treats nuclear deformation explicitly. The strength distributions are then used to calculate the beta-delayed fission, neutron emission, and gamma deexcitation probabilities for these nuclei. Fission is treated both in the complete damping and WKB approximations for penetrabilities through the nuclear potential-energy surface. The resulting fission probabilities difter by factors of 2 to 3 or more from the results of previous calculations using microscopically computed beta-strength distributions around the region of greatest interest for production of the cosmochronometers. The indications are that a consistent treatment of nuclear deformation, fission barriers, and beta-strength functions is important in the calculation of delayed fission probabilities and the production of the actinide cosmochronometers. Since we show that the results are very sensitive to relatively small changes in model assumptions, large chronometric ages for the Galaxy based upon high betadelayed fission probabilities derived from an inconsistent set of nuclear data calculations must be considered quite uncertain.

I. INTRODUCTION

A knowledge of the beta-strength distributions far away from the line of beta stability is crucial for a proper understanding of the astrophysical r process. These strength distributions determine the beta-decay rates of the nuclei involved in the r process. Microscopic calculations of the strength distributions of r-process nuclei have led to improved fits to the observed solar system abundances' compared with fits based on strength distributions computed in the gross theory.² However, even with present microscopically calculated beta-decay rates, an unambiguous solution for the r-process site has not yet been found.³

When coupled with daughter-nucleus fission-barrier heights B_f and neutron-separation energies S_n the beta strength distributions also give the amount of betadelayed fission and beta-delayed neutron emission in the termination of the r-process path and in decay from the r-process path back to the line of beta stability. Because beta-delayed fission may significantly influence the final abundance distribution of elements produced in an rprocess, 3^{-8} it is of vital importance to have reliable predictions of the beta strength function in the calculation of the production ratios of the actinide cosmochronometers.

The actinide production ratios are of interest because when they are compared with observed abundance ratios they can give important information on the galaxy's they can give important information on the galaxy's
age^{8–10} and chemical evolution.^{10,11} Thielemann, Metzinger, and Klapdor⁷ (TMK) used the beta strength distributions of Ref. ¹ to compute new values for the actinide production ratios. The new ratios lead⁷ to an age for the galaxy of (20.8^{+2}_{-4}) Gyr which is significantly greater than previous age estimates from nuclear cosmochronologytypically $13-15$ Gyr.¹² This greater age for the galaxy, if correct, would probably imply¹³ a nonzero cosmological constant in order to yield the Euclidean metric for the universe required by inflationary cosmology.

Recent calculations, however, cast some doubt on the reliability of the results of TMK. Meyer et $al.$ ¹⁴ performed beta-delayed fission and neutron-emission calculations on 115 heavy, neutron-rich nuclei and found less beta-delayed fission than TMK. Their calculations used a beta strength distribution code¹⁵ that treats nuclear deformation explicitly using the random-phase approximation, RPA, to mix a basis of deformed nuclear states via a Gamow-Teller residual interaction. This is to be contrasted with the treatment of Ref. ¹ in which Gamow-Teller matrix elements were calculated with spherical basis states populated so as to give roughly the same occupation numbers for the parent ground state as those from a deformed Nilsson nuclear potential.

This latter approximation omits the extensive mixing of Gamow-Teller strength among deformed states, which is important in determining the beta-decay properties of deformed nuclei. The extent of the effect of this mixing is illustrated in Fig. ¹ which compares the beta strength function for 236 Pa calculated in the present work with

that given in Fig. 4 of TMK. In the present work, the basis of deformed states introduces substantial spreading of the Gamow-Teller strength among the spectrum of daughter states. This will have an important effect on the beta-decay properties of such deformed nuclei since even small amounts of Gamow-Teller strength at low energies in the daughter will give rise to a high decay rate because of the large energy phase-space factor.

Furthermore, in TMK, the fission barriers, the ground-state masses and deformations, and the beta strength functions were obtained from different sources. This can also lead to spurious results, since one must know the location of the beta strength relative to the fission-barrier height. An inconsistent treatment of ground-state masses and fission barriers can lead to artifically high beta strength above the fission barrier and excessive beta-delayed fission. For example, Cowan, Thielemann, and Truran¹⁶ have found that by using a consistent set of masses and fission barriers from Howard and Möller¹⁷ the beta-delayed fission probabilities were dramatically reduced relative to those in TMK. We thus emphasize the importance of computing the ground-state

FIG. 1. Beta strength as a function of energy for 236 Pa. The strength in the upper part of the figure was calculated with our deformed RPA model; the strength in the lower part of the figure was taken from Thielemann, Metzinger, and Klapdor (Ref. 7).

masses, deformations, fission barriers, and beta-strength functions consistently within the same model.

In another recent calculation $H \circ f^{18}$ found that the TMK delayed-fission probabilities were not compatible with thermonuclear explosion mass yields. These findings suggest that a new calculation should be performed of the delayed-fission and neutron-emission probabilities of nuclei of interest to the r process. This new calculation should be internally consistent. That is, the strength distributions, nuclear masses, ground-state deformations, and fission barriers used in the calculation should all be computed from a unified nuclear model that includes an explicit treatment of nuclear deformation.

New models for calculating masses, ground-state deformations, and fission barriers are now becoming available. In particular, recent improvements in the models for the macroscopic energy should give a better description of neutron-rich nuclei than either of the earlier versions of the liquid drop or droplet models. The recent development of a mass formula¹⁹ based on a Yukawa-plusexponential macroscopic model and a folded Yukawa microscopic model gives an improved prediction of the masses of neutron-rich nuclei since the errors in the model do not increase far from β stability (for example, for $99Rb$). The improved properties of the model seem to be due to the inclusion of additional physical effects in the macroscopic energy, such as the finite range of the nuclear force. An improved version of the droplet model has also been proposed. This is the finite-range droplet model²⁰ which combines the earlier droplet model with the folding surface and Coulomb-energy integrals from the finite-range model. It also incorporates a new term that depends on bulk density variations. These effects correct many of the deficiencies of the droplet model. Because these improved models should give more accurate results for the beta-delayed fission and beta-delayed neutron emission probabilities, we find it advisable to defer the large-scale calculation of these probabilities until the necessary quantities are calculated with the new models. For now we present a consistent study of 19 heavy, neutron-rich nuclei in an effort to point out the importance of nuclear deformation in these calculations and to examine the uncertainties resulting from inaccuracies in the fission barriers and in the beta-strength distributions.

II. MODELS

Gamow-Teller beta strength distributions, B_{GT} , were calculated from the RPA code of Krumlinde and Möller,¹⁵ extended by Kratz, Leander, Krumlinde and Möller²¹ to use states in a deformed Woods-Saxon potential. A sample of 19 nuclei in the region of interest for the production of the actinide cosmochronometers was considered. The ground-state deformations used to compute these strength functions were taken from Ref. 17.

The probabilities for fission, neutron emission, and gamma deexcitation following a beta decay are

$$
P_{f} = \frac{\int_{0}^{Q} B_{\text{GT}}(E)f(E)\frac{T_{f}(E)}{T_{\text{tot}}(E)}dE}{\int_{0}^{Q} B_{\text{GT}}(E)f(E)dE} , \qquad (1)
$$

$$
P_n = \frac{\int_0^Q B_{\text{GT}}(E)f(E)\frac{T_n(E)}{T_{\text{tot}}(E)}dE}{\int_0^Q B_{\text{GT}}(E)f(E)dE}, \qquad (2)
$$

$$
P_{\gamma} = \frac{\int_{0}^{Q} B_{\text{GT}}(E)f(E) \frac{T_{\gamma}(E)}{T_{\text{tot}}(E)} dE}{\int_{0}^{Q} B_{\text{GT}}(E)f(E) dE} , \qquad (3)
$$

respectively, where the maximum beta-decay energies Q are taken from Ref. 17; $f(E)$ is the Fermi function; T_f , T_n , and T_γ are the transmission coefficients for fission, neutron emission, and γ decay, respectively; and $T_{\text{tot}} = T_f = T_n + T_\gamma.$

The fission probabilities, P_f , were calculated in two diferent ways. For our best estimates, the transmission coefficients, $T_f(E)$, were calculated for each nucleus by fitting parabolic curves on each side of all extrema in the fission path determined from data in Table I of Ref. 17 and matching the parabolic curves at midpoints between extrema in the fission path. Penetrabilities were then calculated in the WKB approximation using the inertial parameters from Randrup et al.²¹

The $T_n(E)$ were computed by considering emission of a neutron from the nuclear surface at radius R in a direction tangential to the surface (see Ref. 22). The $T_{\gamma}(E)$ were calculated from the model for E1 and M1 transitions found in Holmes et $al.^{23}$. The level-density formula used throughout this work is also that found in Holmes et $al.^{23}$ The level-density at a mass-asymmetric saddlepoint is enhanced by a factor of 2 relative to that for a spherical nucleus because of contributions from the lowenergy rotational states.²⁴ TMK also take this enhancement factor into account.

In order to compare the results of using our strength functions directly with the results of TMK, we also calculated P_f from the complete damping approximation expression.^{7,25} As in TMK, we chose the highest two of the set of axially symmetric, axially asymmetric, and mass asymmetric saddle points as the two barriers through which to penetrate. These barriers were given the curvatures and level-density enhancements prescribed in TMK. The penetrabilities were then computed using the Hill and Wheeler expression²⁶ for parabolic barriers. Because we had no direct simple way of using the same neutronemission strength as TMK, we set $T_n = 0$ throughout. Thus, the P_f values derived for this comparison would be the maximum values TMK would have found using our beta-strength distributions since TMK also calculated T_{γ} via the Holmes et $al.^{23}$ prescription.

The models used here to calculate the branching ratios between fission, neutron emission, and γ deexcitation are standard statistical models. The greatest uncertainty influencing these branching ratios in our model are due to uncertainties in our model for the beta strength function. There are two major contributions to this uncertainty.

First, the accuracy of the calculated Gamow-Teller beta-strength functions depends critically on the accuracy of the level spectrum of the underlying single-particle model. We use for our single-particle model the Woods-

Saxon model discussed in Ref. 27. The incorporation of this model into our calculation of the beta-strength function is discussed in Ref. 21. We use the "universal" parameter set of the Woods-Saxon model. This set has been determined by adjusting calculated single-particle levels to experimental data throughout the periodic system. Although some deviations exist between the calculated and experimental levels, good agreement is obtained in the actinide region. We therefore expect that the calculated beta-strength functions are fairly reliable. However, for the cases where the branching ratios depend critically on the exact position of a single pygmy resonance to within 0.5 MeV or so, the uncertainty in the calculation is still large.

Second, the branching ratios are influenced by the neglect of first-forbidden decay. In general, the effect of neglecting these contributions to the strength function is that the decay rates to low-lying states are underestimated. As we shall see below, nuclear deformation tends to spread out the beta strength function. This leads to more decay to lower-lying states and consequently less betadelayed fission. Forbidden decay would further enhance the decay rates to low-lying states. The role of β -delayed fission would therefore be even smaller if first-forbidden decay were included in the model.

In our calculation we study the decay of even, oddeven, and odd-odd nuclei. In Ref. 15 a model is developed for treating β decay in an RPA approximation with a simple residual Gamow-Teller interaction added to a Nilsson single-particle potential. The model gives most simply the transition amplitudes from the ground state of an even-even nucleus to an odd-odd daughter. To treat the decay from an odd-even parent ground state to an even-odd daughter a perturbation expression was used to generate the odd-particle wave functions. For the odd- A transitions there are two cases. First, there are $\Delta v=0$ transitions from the odd $v=1$ ground state to a $v=1$ state in the daughter nucleus; and second, $\Delta v=2$ transitions to one-quasiparticle phonons, where the initial odd particle only acts as a spectator. The transitions of the second type are the most numerous since they are proportional to the number of nucleons squared, whereas the transitions of the first type are only proportional to the number of nucleons.

Here we have extended the model to the decay of oddodd systems in the following simple approximation. We have used the same perturbation model that was used to generate singly odd systems to generate the odd-odd particle wave function corresponding to the presence of both an odd proton and an odd neutron. We neglect the residual interaction between the odd neutron and odd proton. Most transitions will also in this case be of the $\Delta v=2$ type to a one-quasiparticle phonon in the daughter nucleus, where the two odd-particle orbitals only act as spectators. The $\Delta v=0$ transitions we now treat as in the odd-even case by applying the perturbation expressions derived for the odd-particle wave functions for the oddeven case to one of the odd particles with the other acting as a spectator, and then in a similar way, to the other particle with the first as a spectator. This can be done since we neglected any residual interaction between the odd neutron and proton. One should recall that the number of these transitions is only proportional to the number of nucleons. However, in the other $\Delta v=2$ case where the number of transitions is proportional to the number of nucleons squared, the neglect of this interaction is of little consequence since the coupled neutron and proton in this case act as spectators.

III. RESULTS

In Table I we give the beta Q values, the highest daughter fission-barrier heights, and neutron separation energies for the 19 nuclei studied in this work. Also shown are the results given in TMK using the Howard and Möller¹⁷ masses [see Fig. $5(c)$ of Ref. 7] compared with our P_f 's from the complete damping approximation using our beta strength functions; and our P_f 's, P_n 's, and P_v 's in the Wentzel-Kramers-Brillouin (WKB) barrier penetration calculation. Even in this upper limit with no neutron emission, our fission probabilities are often less than TMK in the complete damping approximation due to the occurrence of more beta strength at low energies in the daughter nucleus (see below). With the WKB barrier-penetration calculations which include competition with neutron emission, we find much less betadelayed fission than TMK, especially in the region around mass number 250. This region is of particular importance for the production of the actinide cosmochronometers since the heaviest α -decaying progenitors of 232 Th and 244 Pu are mass numbers 248 and 252, while the heaviest α -decaying progenitor of ²³⁸U is 250.

Our beta strength functions, which are derived from a fully deformed RPA model calculation, apparently have more strength at lower daughter-nucleus excitation energies than do the strength functions used by TMK, even when the same mass formula and fission barriers are employed. Some of our results are shown in Figs. 2—5. The beta strength function relative to the fission barrier is shown on the left of these figures. The relative betadecay rates to different levels in the daughter nucleus are shown on the right. The present calculations place small amounts of Gamow-Teller strength at low excitation energies in the daughter nuclei due to the mixing of deformed states. Because the strength distributions are weighted by the Fermi function, any strength located at low excitation energy in the daughter will usually cause most of the intensity to be concentrated there. This implies less fission and less neutron emission. Thus, it is evident from this work that the effects of nuclear deformation on beta-strength distributions must be treated as accurately as possible in calculations of delayed fission and neutron emission.

In order to study the effects of uncertainties in fissionbarrier heights, we repeated the calculations, this time with the fission barriers all lowered by ¹ MeV, which is roughly the uncertainty in the fission-barrier estimates. These results are presented in Table II. Substantial increases in the P_f values occur for some cases (e.g., decay of parent nucleus 252 Ac) as expected. Table III shows the results when fission barriers are raised by ¹ MeV. (Note that recent improvements 28 in both the microscopic and macroscopic models discussed here have led to higher

TABLE I. Shown are the beta ^Q values for the decaying parent nucleus, the highest fission barrier $({\bf{B}}_f)$ in the daughter nucleus, and the neutron separation energy $({\bf{S}}_n)$. Also shown are the TMK betadelayed fission probabilities in the complete damping approximation, our beta-delayed fission probabilities in the complete damping approximation, and our beta-delayed fission, neutron emission, and gamma deexcitation probabilities in the WKB barrier penetration treatment.

				Complete damping		WKB barrier penetration		
Parent nucleus	\mathcal{Q}_{β}	B_f	S_n	TMK ^a	This work		This work	
	$(in \text{ MeV})$			P_f	P_f	P_f	P_n	P_{γ}
^{234}Fr	5.07	8.16	5.70	3%	0%	0%	0%	100%
244 Fr	7.90	6.73	4.64	93%	0%	0%	43%	57%
252 Fr	9.51	5.13	3.96	82%	68%	0%	83%	17%
246 Ac	6.79	6.03	4.96	84%	0%	0%	13%	87%
248 Ac	7.23	5.53	4.78	92%	3%	0%	37%	63%
252 Ac	8.20	4.59	4.33	96%	58%	9%	63%	28%
264 Ac	10.30	4.84	3.54	10%	83%	2%	90%	8%
^{250}Pa	6.24	4.80	5.10	89%	8%	10%	1%	89%
$\rm ^{252}Pa$	6.65	4.29	4.90	83%	36%	34%	2%	64%
^{254}Pa	7.28	3.83	4.66	95%	54%	40%	11%	49%
^{260}Pa	8.34	3.82	4.49	100%	92%	97%	0%	3%
270 Pa	10.50	5.15	3.51	2%	81%	14%	77%	9%
252 Np	5.09	4.16	5.42	32%	2%	5%	0%	95%
254 Np	5.56	4.00	5.23	50%	19%	13%	0%	87%
276 Np	10.41	4.46	3.31	25%	83%	9%	84%	7%
$^{251}\mathrm{Am}$	1.90	5.03	4.75	0%	0%	0%	0%	100%
$^{258}\mathrm{Am}$	5.13	4.09	5.29	35%	12%	18%	0%	82%
$^{264}\mathrm{Am}$	6.19	3.28	5.11	100%	85%	100%	0%	0%
$^{277}\mathrm{Am}$	8.04	4.14	2.70	47%	47%	19%	75%	6%

'Using the mass formula of Howard and Moiler {1980).

FIG. 2. Importance of beta-delayed fission in the decay of ²⁴⁴Fr. Nuclear potential energy of ²⁴⁴Ra is shown as a function of distance between mass centers of the nascent fission fragments (in units of 1.16 $A^{1/3}$ fm). On the left (in relative units) is the beta-strength function from the decay of 244 Fr, and on the right (in relative units) are the beta decay-rates to levels in 244 Ra. Notice that the energy phase space factor is important in determining the relative beta-decay rate.

fission barriers for heavy neutron-rich nuclei.) Here we find large decreases in some P_f values (e.g., decay of the parent nucleus 254 Pa) due to the decreased likelihood of penetration through the higher fission barriers. It is clear from the results in Tables II and III that β -delayed fission probabilities can be quite sensitive to uncertainties in the fission-barrier heights.

IV. CONCLUSIONS

It is evident from this work that an accurate knowledge of the shape of the beta strength distribution

FIG. 3. Same as in Fig. 2, except that the parent nucleus is 246 Ac and the daughter nucleus is 246 Th.

FIG. 4. Same as in Fig. 2, except that the parent nucleus is 252 Ac and the daughter nucleus is 252 Th.

is vital to reliable beta-delayed fission calculations. Because the deformation of the decaying nucleus can substantially afFect this shape, calculations of beta-delayed fission and beta-delayed neutron emission probabilities should use beta strength distributions computed from codes that treat nuclear deformation as realistically and consistently as possible. Previous calculations have not done this. The indications from our work are that, when nuclear deformation is treated consistently in the models used to compute beta strength functions, fission-barrier heights, and nuclear masses, the resulting beta-delayed fission probabilities are usually less than previously calculated.

Even with a realistic and accurate treatment of nuclear deformation, however, all calculated beta-delayed fission probabilities, including our own results, are still subject to uncertainties in the nuclear models. For example, as we have seen, the beta-delayed fission and neutron-

FIG. 5. Same as in Fig. 2, except that the parent nucleus is 260 Pa and the daughter nucleus is 260 U.

	Q_{β}	B_f	S_n	Complete damping		WKB barrier penetration		
Parent nucleus				TMK ^a	This work		This work	
	$(in \text{MeV})$			P_f	P_f	P_f	P_n	P_{γ}
^{234}Fr	5.07	7.16	5.70	3%	0%	0%	0%	100%
244 Fr	7.90	5.73	4.64	93%	8%	0%	43%	57%
252 Fr	9.51	4.13	3.96	82%	84%	19%	72%	9%
246 Ac	6.79	5.03	4.96	84%	8%	9%	12%	79%
248 Ac	7.23	4.53	4.78	92%	38%	23%	23%	54%
252 Ac	8.20	3.59	4.33	96%	95%	82%	17%	1%
264 Ac	10.30	3.84	3.54	10%	92%	30%	69%	1%
$^{250}\mathrm{Pa}$	6.24	3.80	5.10	89%	37%	42%	0%	58%
$\rm ^{252}Pa$	6.65	3.29	4.90	83%	59%	61%	0%	39%
254 Pa	7.28	2.83	4.66	95%	81%	80%	0%	20%
260 Pa	8.34	2.82	4.49	100%	97%	97%	0%	3%
270 Pa	10.50	4.15	3.51	2%	99%	33%	67%	0%
252 Np	5.09	3.16	5.42	32%	12%	14%	0%	86%
254 Np	5.56	3.00	5.23	50%	50%	48%	0%	52%
^{276}Np	10.41	3.46	3.31	25%	93%	42%	58%	0%
$^{251}\mathrm{Am}$	1.90	4.03	4.75	0%	0%	0%	0%	100%
258 Am	5.13	3.09	5.29	35%	48%	52%	0%	48%
264 Am	6.19	2.28	5.11	100%	100%	100%	0%	0%
277 Am	8.04	3.14	2.70	47%	89%	59%	39%	2%

TABLE II. The same as Table I but with all fission saddle points lowered by ¹ MeV in our work.

'Using the mass formula of Howard and Moiler (1980).

emission probabilities can be substantially changed by raising or lowering the fission-barrier heights by ¹ MeV, roughly the uncertainty in fission-barrier heights due to uncertainties in the nuclear models used to compute these fission-barrier heights. Thus, until consistent calculations using improved nuclear models are performed, the amount of beta-delayed fission occurring during production of the progeniters of the actinide cosmochronometers remains uncertain. Since our microscopic betastrength functions produce less beta-delayed fission than in previous work,⁷ we conclude that the nuclear cosmochronological age for the galaxy is probably less than that deduced in Ref. 8. However, because of the uncertainties in the properties of heavy neutron-rich nuclei, we also conclude that all present estimates of the cosmochronological age must be considered quite uncertain. More-

TABLE III. The same as Table I but with all fission saddle points raised by ¹ MeV in our work.

				Complete damping		WKB barrier penetration		
Parent	\mathcal{Q}_{β}	B_f	S_n	TMK ^a	This work		This work	
nucleus	$(in \text{ MeV})$			P_f	P_f	P_f	P_n	P_{γ}
234 Fr	5.07	9.16	5.70	3%	0%	0%	0%	100%
244 Fr	7.90	7.73	4.64	93%	0%	0%	43%	57%
252 _{Fr}	9.51	6.13	3.96	82%	23%	0%	83%	17%
246 Ac	6.79	7.03	4.96	84%	0%	0%	13%	87%
248 Ac	7.23	6.53	4.78	62%	0%	0%	37%	63%
^{252}Ac	8.20	5.59	4.33	96%	22%	0%	71%	29%
264 Ac	10.30	5.84	3.54	10%	62%	0%	92%	8%
^{250}Pa	6.24	5.80	5.10	89%	0%	0%	1%	99%
252 Pa	6.65	5.29	4.90	83%	1%	1%	18%	81%
254 Pa	7.28	4.83	4.66	95%	31%	0%	37%	63%
^{260}Pa	8.34	4.82	4.49	100%	44%	94%	0%	6%
^{270}Pa	10.50	6.15	3.51	2%	57%	1%	78%	21%
252 Np	5.09	5.16	5.42	32%	0%	0%	0%	100%
254 Np	5.56	5.00	5.23	50%	1%	1%	0%	99%
276 Np	10.41	5.46	3.31	25%	45%	0%	86%	14%
251 Am	1.90	6.03	4.75	0%	0%	0%	0%	100%
258 Am	5.13	5.09	5.29	35%	0%	0%	0%	100%
264 Am	6.19	4.28	5.11	100%	14%	47%	0%	53%
277 Am	8.04	5.14	2.70	47%	21%	1%	75%	24%

'Using the mass formula of Howard and Moiler (1980).

over, there is no conclusive nuclear cosmochronological evidence for the necessity of a nonzero cosmological constant, as has been proposed.

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