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High resolution neutron capture and transmission measurements on ^{137}Ba and their impact on the interpretation of meteoric barium anomalies

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We have made improved measurements of the neutron capture and total cross sections for ^{137}Ba over a sufficiently wide range of energies so that the reaction rate at *s*-process temperatures ($kT=6$–23 keV) can be determined solely from the data. These rates are crucial for the interpretation of recently discovered anomalies of Ba isotopes in silicon carbide grains from the Murchison meteorite. Recent stellar models of the *s* process are in agreement with the meteoric anomaly data for Ba only if the ^{137}Ba(n,γ) reaction rate is 20% larger than the previously accepted rate. Our reaction rates at *s*-process temperatures are in agreement with the extrapolated reaction rate from the most recent previous measurement. Hence, our results uphold, and place on much firmer footing, the discrepancy between recent stellar models of the *s*-process and the meteoric anomaly data. [S0556-2813(98)50304-4]

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The abundances of the chemical elements and their isotopes in the solar system have arisen from a combination of several different astrophysical sources. For example, there are thought to be three main nucleosynthesis processes which produced the solar system abundances for elements heavier than iron (the so-called *s*-, *r*-, and *p*-processes). The *s*-process path in the region of the element barium is illustrated in Fig. 1. As can be seen, the *s* process contributes to the abundances of the five stable isotopes of barium from ^{134}Ba to ^{138}Ba. The isotopes ^{134}Ba and ^{136}Ba are shielded by stable isobars of xenon against contributions from the *r* process, and the *p* process provides only negligible contributions to their abundances. Hence, these two isotopes of barium are among the relatively few so-called *s*-only isotopes whose abundances are the most important calibration points for models of the *s* process. On the other hand, because they are on the proton-rich side of the valley of beta stability, ^{130}Ba and ^{132}Ba are bypassed by the *s* process and, in addition, they are shielded by stable isobars against contributions from the *r* process. Hence, ^{130}Ba and ^{132}Ba are known as *p*-only isotopes. The three remaining stable isotopes of barium receive contributions from the *r* process, so their average solar system abundances are of limited use for constraining models of the *s* process. However, the discovery of material from certain meteorites with anomalous isotopic composition offers the possibility for disentangling the various nucleosynthesis components, thereby providing additional isotopes which are effectively *s*-only, which can be used to improve nucleosynthesis models.

Microscopic grains of silicon carbide from certain meteorites carry within them trace amounts of several heavy elements having isotopic ratios very different from average solar system material [1–3]. The high precision data from these meteoric SiC grains show linear correlations between the ratios of the abundances of two different isotopes of a given element relative to a third isotope (the so-called three-isotope plots) indicating that the material is a mixture of two components. One component is of almost normal solar composition whereas the second component appears to carry the signature of *s*-process nucleosynthesis. By extrapolating the anomalous meteoric abundance ratios back to zero abundance for *p*-only ^{130,132}Ba [3], the relative *s*-process abundances of the five other stable isotopes of barium were obtained. Because of their apparent *s*-process enrichments and because SiC dust is expected to form in such an environment, it is thought that these grains originated in the circumstellar envelopes of asymptotic giant branch (AGB) stars inside of which the *s* process was occurring [4–6]. As a consequence, data on the isotopic anomalies in these SiC grains can potentially provide a precise test of *s*-process models.

As the data have improved, significant discrepancies have been found between classical *s*-process calculations and both

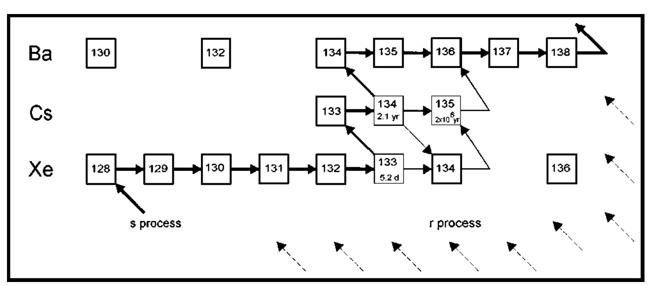

FIG. 1. Nucleosynthesis processes in the Xe-Cs-Ba region. The *s*-process path is depicted by the solid arrows whereas contributions from the *r* process are shown as dashed arrows. The *p* process, is thought to produce the nuclides on the proton-rich side of the valley of stability (such as ^{130,132}Ba) which cannot be reached via the *s* or *r* processes.

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the solar system abundances [7–9] and the meteorite data [10]. In contrast, recent stellar models of the s process occurring in low-mass AGB stars have, for the most part, obtained good agreement with the meteorite data [6] as well as with the overall solar system s-process abundances [5]. One exception to this good agreement occurs for ^{137}Ba. The ^{137}Ba/^{136}Ba s-process ratio inferred from the meteorite data [3] is 20% smaller than the model predictions [6]. The combined uncertainty of the meteorite data and the calculations (whose main uncertainty can be traced to the input neutron capture cross sections) is thought to be about 7%. Furthermore, whereas the isotopic ratios involving nuclides with small neutron capture cross sections can be made to vary considerably by changing the strength of the neutron exposure in the model, the ^{137}Ba/^{136}Ba ratio is only slightly affected by such changes and instead appears to be sensitive only to the ratio of the values used for the neutron capture cross sections (or, more precisely, to the values used for the reaction rates, $N_A\langle\sigma v\rangle$). As a consequence, it has been suggested [6] that the reported ^{137}Ba(n,γ) reaction rate is 20% too small. Similar earlier suggestions [11] about errors in previous determinations for the reaction rates for other isotopes of barium, and for isotopes of neodymium were subsequently verified by recent measurements [7,8,10,12–14].

Although the ^{137}Ba(n,γ) cross section recently has been measured to good precision [12], this measurement is in significant disagreement with the previous data [15]. Also, a major change in the new stellar models of the s process [5] compared to previous models is that there are now two components to each neutron exposure. The major neutron exposure at a temperature of $kT=6-8$ keV is followed by a second and much smaller exposure at $kT=23$ keV. Neither of the two previous ^{137}Ba(n,γ) experiments extended to low enough energies to determine the reaction rate at $kT=6-8$ keV without extrapolating the measured data in some way. For example, the most recent and most precise measurement of the ^{137}Ba(n,γ) cross section [12] had a rather high low energy limit of 10 keV. As a result, the authors [12] estimated that their extrapolation accounted for almost half of the reaction rate at $kT=10$ keV. Furthermore, they estimated that the extrapolation contributes an uncertainty of only 6.9% to the reaction rate at this temperature. However, recent ^{134,136}Ba(n,γ) [8] and ^{116,120}Sn(n,γ) [16] measurements have shown that similar previous extrapolations were in error by as much as 3 times the estimated uncertainties. So, it seemed reasonable to suspect that the ^{137}Ba(n,γ) reaction rate at $kT=6-8$ keV extrapolated from the results of Ref. [12], could be in error by as much as 20%.

For these reasons, we undertook a new measurement of the ^{137}Ba(n,γ) cross section at the Oak Ridge Electron Linear Accelerator (ORELA). Our measurements span a range of energies which is wide enough ($E_n=20$ eV to 280 keV) so that it is not necessary to resort to extrapolations to obtain the reaction rate at the temperatures needed by s-process models. In addition, we improved upon previous measurements by using a thinner sample and also by measuring the ^{137}Ba$+n$ total cross section in a separate experiment. These latter improvements can reduce considerably the systematic uncertainties associated with corrections which must be applied to the ^{137}Ba(n,γ) data because the measurements are necessarily made with samples of finite thickness. Such corrections can be especially important at the low energies needed by the new stellar models.

The general experimental parameters were the same as those in Ref. [8]. The ORELA was operated at a pulse rate of 525 Hz, a pulse width of 8 ns, and a power of 3–5 kW during the course of the experiment. The samples were in the form of isotopically enriched (to 81.72% in ^{137}Ba), compressed barium carbonate powder that was rented from the Oak Ridge Enriched Stable Isotope Pool. The neutron capture and transmission measurements were performed at the same time on different flight paths. We employed the pulse-height weighting technique, using a pair of C_6D_6 scintillators, to make the capture measurements on ORELA flight path 7 at a source-to-sample distance of 40.116 m. The sample for the capture measurement was in the shape of two disks, 2.54 cm in diameter, which were encapsulated in thin-walled aluminum cans and placed one above the other in the neutron beam. The total weight and thickness of the capture sample was 10.0341 g (of $BaCO_3$) and 0.003026 at/b (of Ba), respectively. Separate sample-out background measurements were made using aluminum cans of the same dimensions as the sample holder. In addition, measurements made with a carbon sample were used to subtract the smoothly varying background due to sample scattered neutrons. The overall normalization of the counts to cross section was made via the saturated resonance technique [17] using the 4.9-eV resonance in the ^{197}Au(n,γ) cross section. The transmission data were taken on ORELA flight path 1 at a distance of 79.827 m using a ^{6}Li-loaded glass scintillator. The sample for the transmission measurement had a thickness of 0.01349 at/b. It was encapsulated in a cylindrical copper holder with thin aluminum windows. The $BaCO_3$ sample was cycled with an empty container having the same dimensions as the sample holder and with polyethylene and bismuth absorbers used for background determination.

For $E_n<20$ keV, the resonances were sufficiently well resolved so that they could be fitted using the multilevel R-matrix code SAMMY [18]. Details of the resonance analysis will be published in a subsequent paper. A total of 144 s- and p-wave resonances were fitted. Because of our new transmission data, accurate corrections for the sometimes substantial resonance self-shielding (and smaller multiple-scattering) corrections could be applied on a resonance-by-resonance basis using SAMMY. In the unresolved resonance region ($E_n>20$ keV), the relatively small average corrections for these effects were calculated using the code SESH [19]. The data in this region were also corrected for isotopic impurities using the cross sections of Ref. [8] for ^{134,136}Ba, Ref. [12] for ^{135}Ba, and Ref. [20] for ^{138}Ba. The latest ENDF evaluations [21], normalized to the relevant data set in the region above 100 keV, were used to extend the data of Refs. [12,20] (above 225 keV and 200 keV, respectively) to make this correction. Example plots of the data and the SAMMY fits are shown in Fig. 2.

In Fig. 3, our cross section data have been compressed into the same coarse bins used in previous work and are compared to the data of Refs. [12,15]. Except for the two lowest energy points of Ref. [12], where our results are significantly higher, our data are in good agreement with this most recent previous measurement; hence, there is little doubt that the results of Ref. [15] are systematically low. In

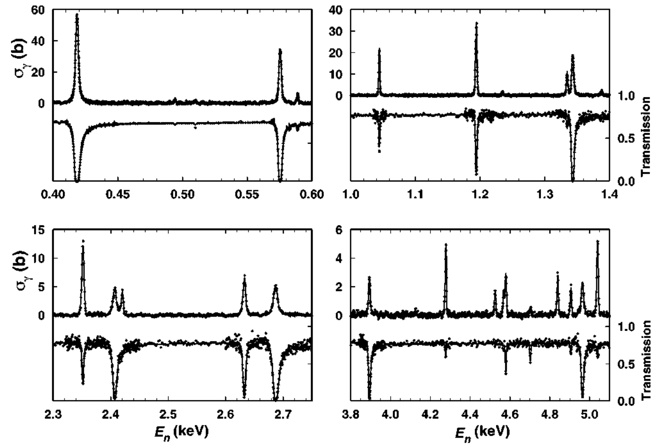

FIG. 2. Representative data (points) and SAMMY fits (solid curves) from our capture (top) and transmission (bottom) measurements on ^{137}Ba. The effective capture cross sections have not been corrected for finite-thickness samples effects. The corrections are included by the code SAMMY; hence, the fits represent the theoretical cross sections, calculated from the resonance parameters, after adjustment for these sample-dependent effects. The scales for the capture data are on the left of each plot whereas the transmission scales are on the right. The transmission data between resonances or over broad resonances were sometimes averaged over several energy bins to reduce the statistical fluctuations. Several of the resonances in the energy region below previous experiments are shown.

addition, contrary to expectations based on our previous measurements [8,16], the extrapolation of Ref. [12] is in good agreement with our data. However, the precision of the reaction rate determined from our data is much better than that of the rate calculated using the extrapolation of Ref. [12]. For example, in Ref. [12], it was estimated that resonances below $E_n=10$ keV contribute 63.13±9.47 mb to the Maxwell-Boltzman averaged cross section at $kT=10$ keV whereas our data show that the actual contribution is 61.4 ±1.8 mb.

The astrophysical reaction rates, $N_A\langle\sigma v\rangle$, calculated from our data are compared to previous results in Fig. 4. In addition, the Maxwell-Boltzman averaged cross sections, $\langle\sigma\rangle$,

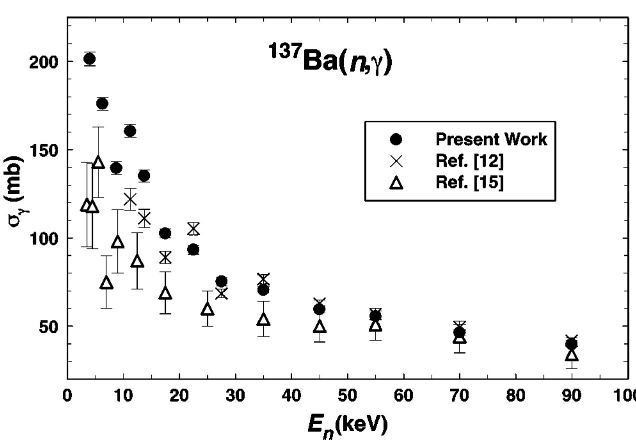

FIG. 3. Comparison between the ^{137}Ba(n,γ) cross sections from the present work (circles) and those of Ref. [12] (X's) and Ref. [15] (triangles). Our data have been compressed into the coarse bins used in Ref. [12].

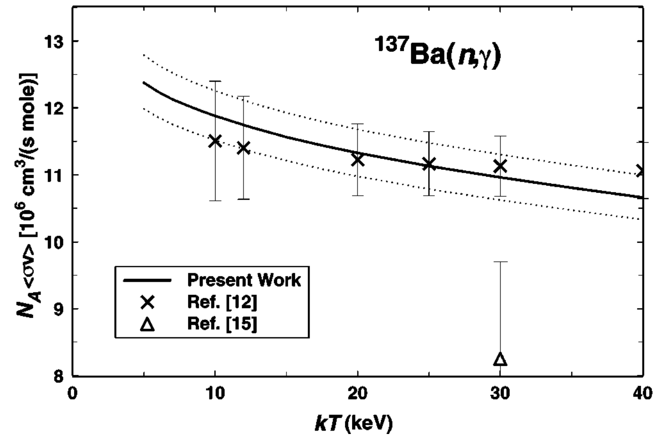

FIG. 4. Astrophysical rates for the ^{137}Ba(n,γ) reaction calculated from the cross sections of the present work (solid curve, with dashed curves depicting the uncertainties), Ref. [12] (X's), and Ref. [15] (triangle).

calculated from our data are given in Table I. The calculated 3% uncertainty in our reaction rates is dominated by contributions from the ^{197}Au(n,γ) and ^{6}Li(n,α) cross sections used to normalize our data. As can be seen in Fig. 4, although the temperature dependence of our reaction rate is somewhat steeper, our results are in agreement with those of Ref. [12] to within the uncertainties. Similarly, a reasonable extrapolation of the results of Ref. [12] from their lowest temperature ($kT=10$ keV) to $kT=6-8$ keV is in agreement with our experimentally determined rates to within the probable uncertainties.

Given the good agreement between our ^{137}Ba(n,γ) reaction rates and those of Ref. [12], and the similarly good agreement between the latest ORELA [8] and Karlsruhe [12] reaction rates for ^{134,136}Ba(n,γ), there can be little doubt that the difference between the stellar s-process models and the meteorite data for ^{137}Ba cannot be attributed to an error in the determination of the (n,γ) reaction rates. At present, no reasonable way to resolve this discrepancy has been identified. However, most of the reaction rates at $kT=6-8$ keV where the bulk of the neutron exposure occurs in the new stellar s-process models are based on extrapolations from data at higher energies. Although the final abundances calculated by these models can be modified by the influence of the second, much smaller exposure at $kT=23$ keV [6,22], accurate determinations of the reaction rates at $kT=6-8$

TABLE I. Maxwellian averaged neutron capture cross sections for ^{137}Ba(n,γ).

kT (keV)	$\langle\sigma\rangle$ (mb) Present Work	Ref. [12]	Ref. [15]
5	209.5 ± 6.8	-	-
8	161.1 ± 5.0	-	-
10	142.2 ± 4.4	137.7 ± 10.6	-
12	128.3 ± 4.0	124.6 ± 8.3	-
20	95.8 ± 3.0	95.0 ± 4.6	-
25	84.2 ± 2.6	84.5 ± 3.7	-
30	75.7 ± 2.4	76.9 ± 3.3	57 ± 10

keV are crucial in many cases. So, more low energy measurements of the type presented herein are needed to more fully test and improve the new stellar *s*-process models and to more fully exploit the meteorite anomaly data for other elements.

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