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NEUTRON CAPTURE CROSS SECTIONS OF CERIUM AND THALLIUM IN THE keV REGION

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Abstract: The neutron capture cross sections of natural cerium and thallium were measured between 10 and 200 keV. The time-of-flight method was used, with a pulsed Van de Graaff accelerator providing the neutron source and with a large liquid scintillator detector. The measured cross sections are extremely low owing to the near-magic nucleon numbers of Ce and Tl. They were averaged over Maxwellian velocity distributions with kT values between 10 and 100 keV so as to be useful for calculations of nucleosynthesis in stars.

E | NUCLEAR REACTIONS Ce, Tl(n, γ), E = 10-200 keV; measured $\sigma(E)$; deduced averages over Maxwellian velocity distributions. Time-of-flight method.

1. Introduction

In the last decade fast-neutron capture cross sections of many elements were measured, often to obtain data for calculations for fast breeder reactors, particularly on elements with large capture cross sections. However, capture data on elements with magic or near-magic nucleon numbers are still scarce. These elements have very small capture cross sections ^{1}) which are not easy to measure with present-day neutron sources. Just because of their "magic" properties, however, they are of special interest in nuclear physics ^{2}) and perhaps even more so in astrophysics.

According to modern concepts of element formation ^{3}) a large fraction of the existing elements heavier than iron was formed by relatively slow successive neutron capture in the interior of stars. The temperatures for this "s-process" ^{3}) are thought to correspond to roughly 20 to 50 keV. Attempts are being made to reproduce the observed solar abundances in detailed calculations using the s-process concept. Input data are the seed abundances and capture cross sections averaged over Maxwellian speed distributions for the assumed stellar temperatures ^{4}). Unknown cross sections are usually estimated by means of the rule that the product of observed abundance and averaged capture cross section varies only slightly from element to element [for details see ref. ^{4})]. At present, however, it is still difficult to say how good such estimates are, especially since solar abundances are known only with large uncertainties.

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For nucleosynthesis calculations two key elements are Ce (two main isotopes with 82 and 82+2 neutrons) and Tl (two main isotopes with 82-1 protons and 126-2 resp. 126-4 neutrons) [ref. [5])]. Both elements have insufficiently known cross sections. Moreover, cerium is one of the more abundant fission products and therefore its fast-neutron capture cross section has also a certain technological importance. In the present paper we present neutron capture cross sections for these elements between 10 and 200 keV neutron energy. In addition we list Maxwellian averages for kT values between 10 and 100 keV calculated with the measured data.

2. Experimental procedure

The apparatus and the experimental procedure employed have been reported earlier [6]), therefore we shall give only a short description here. The measurements were performed by the time-of-flight method at a pulsed 3 MV Van de Graaff accelerator, with 10 ns pulses and a 1.5 m flight path. Neutron capture events were detected in an 800 l liquid scintillator surrounding the sample in 4π geometry. The capture cross sections were measured relative to gold.

The samples were metallic discs of 8.6 cm diameter containing 163 g of natural cerium ($1.207 \cdot 10^{-2}$ nuclei/b) with a purity of 99.9 % and 128 g of natural thallium ($0.650 \cdot 10^{-2}$ nuclei/b) with a purity of 99.999 %. Each of these samples was interchanged at short time intervals with a gold sample which provided the standard and with a graphite scatterer which served to determine the time-dependent background caused by neutrons scattered by the sample and subsequently captured in the structural materials of the detector. The thickness of the graphite scatterer in terms of mean free paths was chosen as close as possible to that of the sample under study. With the very low capture cross sections investigated here this background determination was crucial.

3. Results

3.1. MEASURED DATA AND ERROR ESTIMATES

The results of our capture measurements are shown in figs. 1 and 2. Above 20 keV the spacing of successive data points corresponds roughly to the experimental resolution. Below 20 keV channels were grouped in the time-of-flight spectra in order to improve the statistical accuracy. Although we did not attempt to resolve resonances it can be seen that our resolution was good enough to reveal pronounced resonance structure for both elements, especially at low energies. The contributions to the experimental uncertainties are estimated as follows.

(i) Our reference cross section is the (n, γ) cross section of gold as measured by Kompe against ^{10}B(n, $\alpha\gamma$) and ^{6}Li(n, α) [cf. refs. [6,7])], normalized to the absolute value 596 mb at 30 keV recommended by Poenitz [8]). Linac measurements recently published by Fricke and Lopez [9]) and a reanalysis of shell transmission data by Semler [10]) indicate that our reference cross section may be too low. The uncertainties

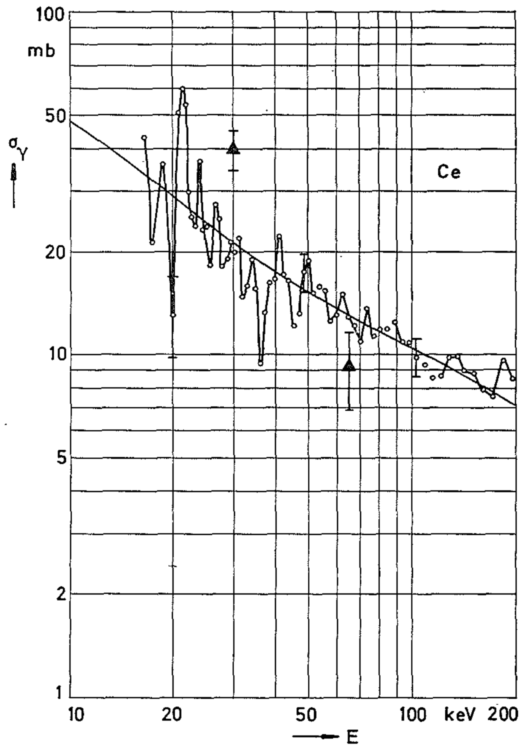

Fig. 1. Cerium capture cross-section data. The open circles are the result of this work, the triangles are the data of Gibbons *et al.* [16]). The line connecting the experimental points is only a guide for the eye, the smooth curve is a χ^2 fit to our data with $S_0 = 0.9 \cdot 10^{-4}$ (fixed parameter), $S_1 = (0.79 \pm 2.1) \cdot 10^{-4}$, $S_2 = (0.16 \pm 0.25) \cdot 10^{-4}$, $S_\gamma = (0.17 \pm 0.07) \cdot 10^{-4}$ (adjusted parameters). All data correspond to the same gold standard cross section.

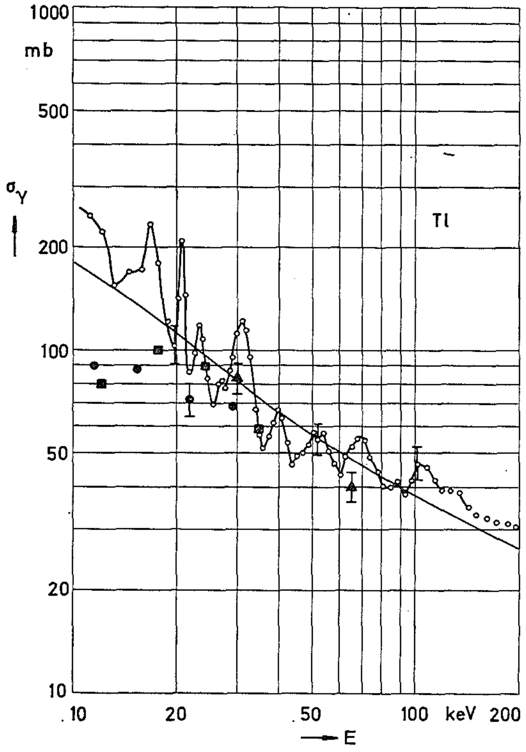

Fig. 2. Thallium capture cross sections. The open circles are the result of this work, the triangles are the data of Gibbons *et al.* [16]), the solid circles are data reported by Konks and Shapiro [17]) for natural thallium. The solid squares are data calculated by Konks and Shapiro [17]) from capture data measured with thin enriched samples of ^{203}Tl and ^{205}Tl. The line connecting the data points is a mere eye-guide, the smooth curve is a χ^2 fit to the data with $S_0 = 1.2 \cdot 10^{-4}$, $S_1 = 1.3 \cdot 10^{-4}$ (fixed parameters), $S_2 = (0.18 \pm 0.11) \cdot 10^{-4}$, $S_\gamma = (0.94 \pm 0.06) \cdot 10^{-4}$ (adjusted parameters). All data correspond to the same gold standard cross section.

given in ref. [8]), 3 % near the calibration point at 30 keV, increasing to 8 % at 200 keV, were therefore raised here to 8 % and 13 %, respectively.

(ii) The uncertainty in the probability for interaction of at least one photon out of a capture γ-ray cascade in the scintillator is estimated as 3 % [see ref. [6])].

(iii) The uncertainty in the probability that a detector signal from a capture event exceeds the discriminator threshold (the uncertainty in the spectrum fraction) is estimated as 6 %.

(iv) Corrections for self-shielding and multiple scattering were calculated by the method proposed by Macklin [11]) based on work by Dresner [12]). This method is supposed to be adequate if each data point represents an average over many resonances. This is obviously not the case here, but since a rigorous calculation requires an iterative resonance analysis and since the corrections were never larger than about 10 % we considered the Macklin formulae as applicable, with an estimated error of 3 %.

(v) The statistical error in the corrected data (ratios of background-corrected counting rates) is listed for three representative energies in table 1.

TABLE 1

Energy (keV)	Statistical uncertainty for each data point (%)		Total uncertainty for each data point (%)	
	Ce	Tl	Ce	Tl
20	11	8	16	14
50	7	5	13	12
100	5	4	14	13

From these contributions one obtains the total uncertainty estimates listed in table 1, and indicated by error bars in figs. 1 and 2.

3.2. MAXWELLIAN AVERAGES

A large fraction of the existing heavy elements is thought to stem from neutron capture in stars which have burnt most of their hydrogen [3]). The interior of these stars is assumed to contain mainly helium (density $\approx 10^4$ g/cm^3) and an extremely small amount of heavy nuclei at a temperature of about $3 \cdot 10^8$ °K. Under these conditions neutrons originating in charged-particle reactions will be thermalized before being captured by the heavy nuclei. This means that the probability for the speed of a neutron relative to the capturing nucleus is given by the Maxwell-Boltzmann distribution

$$p(v, T)\mathrm{d}v = \frac{4}{\sqrt{\pi}} \left(\frac{v}{v_\mathrm{T}}\right)^2 \exp\left[-\left(\frac{v}{v_\mathrm{T}}\right)^2\right] \frac{\mathrm{d}v}{v_\mathrm{T}}, \qquad (1)$$

where v_T is the most probable relative speed ($\frac{1}{2}mv_\mathrm{T}^2 = kT$, m is the reduced mass, and kT the temperature in energy units). The capture rate per unit volume for a given seed nucleide with capture cross section σ_γ is

$$Nn\langle v\sigma_\gamma\rangle \equiv N\langle nv\rangle\langle\sigma_\gamma\rangle_{\mathrm{T}}, \tag{2}$$

where N and n are the densities of seed nuclei and neutrons, respectively, $\langle\ldots\rangle$ denotes an average over the Maxwell-Boltzmann distribution (1), and $\langle\sigma_\gamma\rangle_{\mathrm{T}}$ is the Maxwellian average mentioned above. Since nv is the relative flux, $\langle\sigma_\gamma\rangle_{\mathrm{T}}$ is an average over the flux distribution,

$$\langle\sigma_\gamma\rangle_{\mathrm{T}} = \frac{\langle v\sigma_\gamma\rangle}{\langle v\rangle} = \frac{\int_0^\infty \mathrm{d}vp(v, T)v\sigma_\gamma(v)}{\int_0^\infty \mathrm{d}vp(v, T)v}. \tag{3}$$

In order to calculate $\langle\sigma_\gamma\rangle_{\mathrm{T}}$ one has to know σ_γ over a rather wide range of energies. Our measurement covered the range between 10 and 200 keV which gives the main contribution to $\langle\sigma_\gamma\rangle_{\mathrm{T}}$ at the stellar temperatures considered here. The contributions from beyond this region were estimated as follows: statistical theory yields for the average capture cross section, i.e. for the average over many unresolved, non-overlapping resonances with spins J, parities $(-1)^l$, spin factors g_J [see refs. [6,13]]

$$\bar{\sigma}_\gamma = 2\pi^2 \lambda\!\!\!^{-2} \sum_{l,J} g_J \frac{\varepsilon_J^{Il} S_l v_l \sqrt{(E/1\ \mathrm{eV})} S_\gamma f(J)}{\sum_{l'} \varepsilon_J^{I'l'} S_{l'}\, v_{l'} \sqrt{(E/1\ \mathrm{eV})} + S_\gamma f(J)}. \tag{4}$$

TABLE 2

Maxwellian averages [a]

kT (keV)	Ce	$\langle\sigma_\gamma\rangle_{\mathrm{T}}$ [b] (mb)	Tl
10	41 ±10		185±40
15	31 ± 6		135±30
20	26 ± 5		111±20
25	22 ± 3		94±11
30	19.9± 3.2		83± 9
35	18.1± 3.0		74± 8
40	16.6± 2.5		68± 7
45	15.5± 2.0		63± 6
50	14.5± 1.6		59± 6
60	13.1± 1.5		53± 5
70	11.9± 1.4		48± 5
80	11.0± 1.3		44± 4
90	10.2± 1.2		41± 4
100	9.6± 1.2		38± 4

[a] It should be noted that the quantity $\langle v\sigma_\gamma\rangle/v_{\mathrm{T}}$ introduced by Macklin and Gibbons [18]) exceeds the average $\langle\sigma_\gamma\rangle_{\mathrm{T}}$ listed here by a factor $2/\sqrt{\pi} \approx 1.13$.

[b] See eq. (3). The uncertainties quoted are due to cross section uncertainties (compare table 1) and extrapolation uncertainties (taken as one third the contribution from the extrapolation).

We used here conventional notation: $f(J) \approx (2J+1)e^{-J(J+1)/2\sigma^2}$ describes the energy dependence of the level density according to $D_J = D/f(J)$, and $S_\gamma \equiv \Gamma_\gamma/D$ with Γ_γ taken as independent of l and J. Inelastic scattering does not occur below 200 keV for the elements investigated here.

A least-squares fit of (4) to the data was made by adjusting the strength functions S_1, S_2, S_γ for cerium, and S_2, S_γ for thallium. Partial waves with $l > 2$ were neglected. The values of S_0 were taken from Seth's compilation [14]), S_1 for Tl from ref. [15]). The best fits obtained in this way are shown in figs. 1 and 2 as solid lines. They were used to extrapolate $\bar{\sigma}_\gamma$ into the regions below and above the range of our measurement. The strength functions fitting our data are given in the captions of figs. 1 and 2. They should not be taken too seriously, however. Although perfectly adequate for the calculation of the Maxwellian average $\langle\sigma_\gamma\rangle_T$ (contributions from outside the range of our measurement are smaller than 5 % except at the lowest temperatures) the strength functions found in the fit have large uncertainties.

The calculated Maxwellian averages and their estimated uncertainties are listed in table 2.

4. Discussion

The only available data on neutron capture by natural cerium and thallium in the keV region are the cerium and thallium cross sections at 30 and 65 keV reported by Gibbons *et al.* [16]) and the thallium cross sections published by Konks and Shapiro [ref. [17])]. The former data were obtained relative to indium, with reference values of 763 mb at 30 keV and 448 mb at 65 keV. Using the same standard cross section the authors measured a gold capture cross section of 515 mb at 30 keV and 332 mb at 65 keV. Renormalizing their data to the gold cross section adopted here (596 mb at 30 keV) one obtains the values shown in figs. 1 and 2 as solid triangles. For thallium there is agreement with our data within the error bars. The cerium values, however, differ significantly, the 30 keV cross section exceeding our data by almost a factor of two, and the 65 keV value being much lower. An indication that the Oak Ridge values may have larger errors than estimated by the authors could be seen in the fact that they are not compatible with the $1/E$ behaviour of $\overline{\sigma_\gamma}$ which one expects qualitatively in the keV region [cf. ref. [6])].

The Russian values were measured with the slowing-down-time spectrometer (lead pile) relative to low-energy resonances in Br, Ta and Au. They are lower than our data. Two samples were used, and a consistent discrepancy exists between the two resulting sets of data. The thicker sample yielded smaller cross sections which may be an indication that resonance self-shielding was underestimated. This hypothesis appears to be confirmed by the fact that values which Konks and Shapiro calculated from data taken with very thin enriched samples of ^{203}Tl and ^{205}Tl [also plotted in ref. [17])] agree much better with our data.

Our Maxwellian averages for $kT = 30$ keV can be compared to the values used by Amiet and Zeh in their s-process calculations. These authors estimate $\langle v\sigma_\gamma\rangle/v_T$ as

12 mb for ^{140}Ce and 200 mb for ^{142}Ce, which corresponds to $\langle\sigma_\gamma\rangle_T = 29$ mb for natural cerium (see footnote to table 2). This is considerably larger than our value $\langle\sigma_\gamma\rangle_T = (20\pm2)$ mb. For Tl there is no big discrepancy. The values 140 mb for ^{203}Tl and 50 mb for ^{205}Tl correspond to $\langle\sigma_\gamma\rangle_T = 68$ mb for natural thallium. Our value of (83 ± 10) mb agrees reasonably well with this estimate.

5. Conclusion

The very low neutron-capture cross sections of the near-magic elements cerium and thallium were measured between 10 and 200 keV. They show considerable resonance structure. The steep decrease of the cerium cross section reported in ref. ^{16}) could not be confirmed. Our Maxwellian average at $kT = 30$ keV for cerium is only two thirds of the value estimated in ref. ^{4}) on the basis of the rule "abundance times capture cross section is a smooth function of mass number". If this rule is valid one must conclude that the solar abundance of cerium should be 50 % higher than the presently known value.

The thallium data are in reasonable agreement with the few previously measured values and with the estimates used in nucleosynthesis calculations.

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