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Re-examination of 2200 metre/second cross section experiments for NEUTRON CAPTURE AND FISSION STANDARDS

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Re-examination of 2200 metre/second cross section experiments for neutron capture and fission standards

Abstract - The measurements of absolute standards for neutron induced capture reactions and neutron induced fission reactions at thermal neutron energies have been compiled and reevaluated. These standards include capture reactions in ^{197}Au , and ^{59}Co , and fission reactions in ^{235}U and ^{239}Pu . The recommended thermal neutron cross section values for these reactions are as follows: $\sigma_{\gamma}(^{197}\text{Au}) = 98.7 \pm 0.1$ barn; $\sigma_{\gamma}(^{59}\text{Co}) = 37.19 \pm 0.08$ barn; $\sigma_f(^{235}\text{U}) = 586. \pm 2.$ barn; and $\sigma_f(^{239}\text{Pu}) = 752. \pm 3.$ barn.

INTRODUCTION

Neutron capture and fission reaction cross sections are usually measured on a relative basis, i.e. a ratio measurement based on some standard value. The most common absolute cross section standards used in the thermal neutron energy region are capture reactions in ^{197}Au , and in ^{59}Co and the fission reactions in ^{235}U and in ^{239}Pu .

In the epi-thermal energy region, i.e., between about 1 electron-volt, (eV)^a, and a few keV, especially for elements with medium to high mass numbers, there are often particular energies for which the rate of neutron capture or fission is exceptionally large. These narrow peaks or resonances in the cross section curve have an analytical form given by the dispersion formula analogous to optical dispersion or a resonating electrical circuit, i.e.:

$$\sigma_{\gamma} = \sigma_0 \left(\Gamma_{\gamma} / \Gamma \right) \left(E_0 / E \right)^{1/2} \left(\Gamma^2 / [\Gamma^2 + 4(E - E_0)^2] \right) \quad (1)$$

and

$$\sigma_0 = 4 \pi \lambda^2 g \Gamma_n / \Gamma = (2.604 \times 10^6 / E(\text{eV})) [A+1/A]^2 g \Gamma_n / \Gamma, \quad (2)$$

where the resonance parameters are defined as follows: E_0 , σ_0 are the energy and the peak cross section of the resonance, λ is the neutron wave length, g is the statistical weight factor of the spin of the target nucleus, A is the mass number of the target nucleus, and Γ , Γ_{γ} , Γ_n are the total, radiation and neutron scattering widths at half maximum of the resonance. For the fission cross section, the fission width, Γ_f , would be substituted for the capture width, Γ_{γ} , in the first equation. If there are no nearby resonances, the cross section varies as $(1/E)^{1/2}$ (i.e. a $1/v$ dependence), where v is the neutron velocity. A non- $1/v$ dependence can be calculated from the (Breit-Wigner) dispersion formula above.

In the following sections, the thermal neutron capture and fission cross sections will be determined from measurements performed at a neutron velocity of 2200 meter/second (which corresponds to neutron energies at room temperature, i.e., about 1/40 of an electron volt or more precisely, 0.0253 eV). It might be noted that there are two other elements which had previously been used as thermal neutron cross section standards, i.e., boron and lithium. In both elements, the large cross section for the natural element is due to a specific reaction in a particular isotope ^6Li or ^{10}B . However, a known variation in the isotopic composition of ^6Li and ^{10}B in nature makes these elements poor choices as standards, since the cross section for the natural element varies significantly depending on the source of the sample. In addition, depleted lithium with only about 1/3 of the normal isotopic abundance of ^6Li and thus only about 1/3 of the usual thermal neutron cross section has been known to have found its way into commercial lithium stocks, see de Goeij (ref. 1).

^a(1 electron-volt = 1.602×10^{-19} joules)

GOLD CAPTURE CROSS SECTION STANDARD VALUE

The most common reference standard for neutron capture reactions is the gold cross section. In this study, no measurements were considered which were made relative to another cross section. In addition, only measurements in which the neutron energy was measured with high resolution and accuracy were considered. This eliminated uncertainties in the determination of the shape of the neutron flux incident on the standard. Questions as to whether a true Maxwellian shape existed for the neutron distribution, and what was the actual temperature of this distribution were also avoided by this restriction. Errors caused by any epi-thermal component of the neutron distribution, which could contribute significantly due to the very large resonance in gold at the energy of 4.9 eV were similarly eliminated.

Table 1 lists all of the experiments which satisfied the above criteria. The total cross section was measured below the Bragg scattering cutoff, where scattering contributions to the total cross section become small (about 0.1%). The $1/v$ curve is fitted in this long wavelength region and extrapolated to a neutron velocity of 2200 meter/second (m/s). A correction for the non- $1/v$ portion of the gold cross section due to the 4.9 eV resonance is made by using the parameters of this resonance (see ref. 2). This non $1/v$ portion contributes 0.902 ± 0.025 barn to the value of the cross section at 2200 m/s.

TABLE 1. $^{197}\text{Au}(n,\gamma)$ cross section measurements

Reference Author (Year)	Reported (barn ^b)	Revised (barn ^b)	Comments
Carter ³ (1953)	98.7 \pm 0.6	98.72 \pm 0.48	Corrected non- $1/v$ portion
Egelstaff ⁴ (1954)	98.6 \pm 0.9	98.6 \pm 0.9	
Gould ⁵ (1960)	98.8 \pm 0.3	98.7 \pm 0.33	Corrected non- $1/v$ portion
Teutsch ⁶ (1962)	98.9 \pm 0.3	98.8 \pm 0.33	Corrected non- $1/v$ portion
Als Nielsen ⁷ (1964)	98.6 \pm 0.2	98.58 \pm 0.25	Corrected non- $1/v$ portion
Dilg ⁸ (1973)	98.68 \pm 0.12	98.63 \pm 0.14	Corrected non- $1/v$ portion
Weighted Average = 98.67 barn ^b \pm 0.10 (Internal Error), \pm 0.03 (External Error).			
Recommended Value = 98.7 \pm 0.1 barn ^b .			

^b(1 barn = 10^{-28} m²)

COBALT CAPTURE CROSS SECTION STANDARD VALUE

For neutron capture reactions, cobalt is used as a standard more often than any other material with the exception of gold. In addition to the ground state of 5.27 years, there is also an isomer at 58.6 keV excitation energy, which has a half-life of 10.5 minutes. The isomeric state decays primarily to the ground state by conversion electron and γ -ray emission. However, there are two weak β branches of 0.24% and 0.0086% intensity. When ^{60}Co is formed in thermal neutron capture, 56% of the compound nuclei are formed in the isomeric state. Activation of the 5.27 year state does not account for all neutron absorption in ^{59}Co . As a result, the cobalt activation cross section is 0.139% less than the cross section for absorption; i.e. a 0.052 barn difference. The quoted cross section following in Table 2 should be reduced by 0.05 barn to obtain the activation value; i.e. 37.14 barn. Table 3 gives the activation value to the 10.5 minute isomeric state.

In Tables 2 and 3, the cross section values as reported by the authors have been revised to renormalize the measurement for the above recommended cross section value for the gold standard (see Table 1), to correct the assumed half-life to the values mentioned above, to correct the γ -ray energy and the γ -ray intensity, $E_\gamma(I_\gamma)$, to the latest recommended values, and to correct the Westcott factor, $g(\sigma)$, which accounts for both spectrum and temperature effects (ref. 9). When the isomeric cross section, σ^m , is stated as a ratio to the ground state cross section, σ^g , σ^g is normalized to the recommendation of Table 2.

TABLE 2. $^{59}\text{Co}(n,\gamma)$ cross section measurements

Reference Author (Year)	Reported (barn ^b)	Revised (barn ^b)	Comments
Vaninbrouckx ¹⁰ (1968)	37.4 ± 0.3	37.3 ± 0.34	Revised $g(\sigma)$, $T_{1/2}$, gold σ , revised uncertainty
Merritt ¹¹ (1968)	37.09 ± 0.27	37.16 ± 0.28	Revised $g(\sigma)$, $T_{1/2}$, gold σ , revised uncertainty
Kim ¹² (1968)	36.61 ± 0.47	37.19 ± 0.52	Revised $T_{1/2}$, gold σ , $E_{\gamma}(I_{\gamma})$, revised uncertainty
Silk ¹³ (1970)	37.245 ± 0.11	37.245 ± 0.15	Revised uncertainty
Dilg ⁸ (1973)	37.145 ± 0.07	37.145 ± 0.11	Revised uncertainty
Gryntakis ¹⁴ (1978)	37.45 ± 0.45	37.45 ± 0.49	Revised uncertainty
Weighted Average = 37.19 barn ^b ± 0.08 (Internal Error), ± 0.03 (External Error).			
Recommended Value = 37.19 ± 0.08 barn ^b .			

^b(1 barn = 10⁻²⁸ m²)

TABLE 3. $^{59}\text{Co}(n,\gamma)^{60m}\text{Co}$ cross section measurements

Reference Author (Year)	Reported (barn ^b)	Revised (barn ^b)	Comments
Deutsch ¹⁵ (1951)	$\sigma^g/\sigma^m = 1.4 \pm 0.6$	15.2 ± 6.5	Revised both half-lives, used recommended σ^g
Moss ¹⁶ (1953)	18.3 ± 1.7 barn	21.8 ± 2.0	Revised gold standard, revised beta branching ratio
Keisch ¹⁷ (1963)	$\sigma^m/\sigma^g = 1.19 \pm 0.16$	21.8 ± 1.4	Revised total conversion coefficient - (tcc)
Schmidt-Ott ¹⁸ (1963)	16.5 barn	17.8 ± (2.0)	Revised (tcc), half-life, revised copper standard
Gryntakis ¹⁴ (1978)	18.8 ± 1.5 barn	19.7 ± 1.6	Revised $g(\sigma)$, gold standard, revised $E_{\gamma}(I_{\gamma})$
Arifov ¹⁹ (1978)	$\sigma^m/\sigma^{m+g} = 0.57 \pm 0.03$	21.2 ± 1.1	Used recommended σ^g
Weighted Average = 20.7 barn ^b ± 0.67 (Internal Error), ± 0.38 (External Error).			
Recommended Value = 20.7 ± 0.7 barn ^b .			

^b(1 barn = 10⁻²⁸ m²)

²³⁵U FISSION CROSS SECTION STANDARD VALUE

The most common standard for the measurement of neutron induced fission reaction cross sections is ^{235}U . There have been many measurements performed in the past forty years, but much of the early data has had to be discarded because auxiliary information on details of the measurements have been unavailable. The various measurements of the direct 2200 meter/second cross section have been evaluated and tabulated in Table 4. Measurements of the cross section in a Maxwellian flux have not been considered here because of the difficulties involved with estimation of the magnitude and shape of the neutron flux in those measurements. The assumption that the flux is a perfect Maxwellian and that the temperature is perfectly known leads to equivalent 2200 m/s cross section values for those experiments, which are significantly smaller by five or more standard deviations than the direct 2200 m/s fission cross section measurement values.

The values listed in Table 4 were renormalized using an absorption cross section for ^{10}B of 3838 ± 6 barn (ref. 2), and a branching ratio, to the excited state of ^7Li , for the $^{10}\text{B}(n,\alpha)$ reaction of $(93.7 \pm 0.1)\%$. The half-life of ^{234}U was assumed to be $2.455 \pm 0.006 \times 10^5$ years (ref. 20), in order to determine the amount of fissile material.

TABLE 4. $^{235}\text{U}(n,f)$ cross section measurements

Reference Author (Year)	Reported (barn ^b)	Revised (barn ^b)	Comments
Saplakoglu ²¹ (1958)	605. ± 6.	590.5 ± 15.8	Zero sample thickness extrapolation correction
Deruytter ²² (1961)	587. ± 6.	590. ± 8.	Corrected scattering in gold foil
Maslin ²³ (1965)	572. ± 6.	581.4 ± 9.4	Zero sample thickness extrapolation correction
Borcea ²⁴ (1973)	581.7 ± 7.8	587.3 ± 7.9	Revised half-life, boron cross section, revised boron branching, correction for fission losses
Deruytter ²⁵ (1973)	587.6 ± 2.6	585.6 ± 3.0	Revised half-life, boron cross section
Berceanu ²⁶ (1978)	589.3 ± 4.7	586.5 ± 5.3	Revised boron branching, revised amount of fissile material
Weighted Average = 585.8 barn ^b ± 1.6 (Internal Error), ± 0.6 (External Error).			
Recommended Value = 586. ± 2. barn ^b .			

^b(1 barn = 10⁻²⁸ m²)

^{239}Pu FISSION CROSS SECTION STANDARD VALUE

In addition to its importance as a fuel material in the nuclear power reactor program, the fission cross section of ^{239}Pu is also of interest as a standard. It is second only to ^{235}U in its use as a fission cross section standard. As in the case of ^{235}U , there are only a few measurements which meet the criteria of a direct determination at the neutron velocity of 2200 meter/second and these values are reported in Table 5. There has been a problem with experiments on ^{239}Pu because the determination of the amount of material in the sample is made using α counting and the α half-life. This half-life has been decreased by more than 1.2 % over the past 10 to 15 years. In addition, whereas this half-life had been thought to be accurate to 0.1% at the 3 standard deviation level (99.7% confidence level), the uncertainty is now estimated to be about 0.15% at the 1 standard deviation level (68.3% confidence level). In Table 5, the half-life uncertainty is treated as a systematic error. The half-life value, which has been used for the renormalization of the amount of fissile material, is $2.410 \pm 0.003 \times 10^4$ years (ref. 20). The values used for the boron absorption cross section and the boron branching ratio are the same as in the previous section.

TABLE 5. $^{239}\text{Pu}(n,f)$ cross section measurements

Reference Author (Year)	Reported (barn ^b)	Revised (barn ^b)	Comments
Borcea ²⁷ (1970)	741.0 ± 7.0	752.9 ± 7.6	Revised half-life, boron cross section, revised boron branching, correction for fission losses
Deruytter ²⁸ (1974)	741.9 ± 3.4	751.5 ± 3.6	Revised half-life, boron cross section
Weighted Average = 751.8 barn ^b ± 3.3 (Internal Error). ± 0.2 (External Error).			
Recommended Value = 752. ± 3. barn ^b .			

^b(1 barn = 10⁻²⁸ m²)

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