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Evaluation of the Thermal-Neutron Constants for ^{233}U, ^{235}U, ^{239}Pu and ^{241}Pu*

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A consistent set of best values of the 2200 meter/second neutron cross sections, Westcott g-factors, and fission neutron yields for ^{233}U, ^{235}U, ^{239}Pu and ^{241}Pu are presented.

A least squares fitting program, LSF, is used to obtain the best fit and to estimate the sensitivity of these fissile parameters to the quoted uncertainties in experimental data.

The half-lives of the uranium and plutonium nuclides have been evaluated and these have been used to reassess the significant experimental data. The latest revision of the spontaneous fission neutron yield $\bar{\nu}$, of ^{252}Cf and the foil thickness corrections to the fission neutron yield ratios of fissile nuclei to ^{252}Cf are included. These lead to greater consistency in the data used for $\bar{\nu}$ (^{252}Cf). Similarly, the ^{234}U half-life as revised leads to improved consistency in the ^{235}U fission cross section.

Comparison is made with the values from ENDF/B-V and other evaluations.

Introduction

The present work, like the efforts that preceded it in 1965,[1] 1969,[2] 1974,[3] 1975,[4] and 1977[5] analyzes and combines all the relevant experimental measurements that lead to a knowledge of the thermal neutron constants for the four principal fissile isotopes: the absorption, fission, and scattering cross sections, and the prompt and total nubar values for thermal neutron fission. Measurements of nubar for ^{252}Cf are included as well. The results of both absolute and relative measurements are combined by the same iterative least-squares fitting program LSF[6] that was used in previous efforts.

The analysis involves adjusting older data to be consistent with current values of the standard neutron cross sections and of the half-lives of the fissile materials shown in Table 5.

The Westcott g-factors have been updated by using the values proposed by Leonard[7,8] for ^{235}U and ^{239}Pu, and those of ENDF/B-V for ^{233}U and ^{241}Pu. Westcott's[9] original uncertainty values are used except for fission in ^{235}U and ^{239}Pu, where Leonard's uncertainty values are quoted. We simplify the interpretation of the old scattering and total cross section measurements by ignoring the small differences in scattering that used to be attributed to crystal structure effects (e.g., metal vs. liquid). We also simplify the calculation of corrections for neutron detector sensitivity in measurements of prompt nubar ratios by adopting the mean fission neutron spectrum energies given by A. B. Smith[10], rather than treating these means as parameters to be adjusted in the LSF fitting (we find the results of the fit to be insensitive to the set of mean energies chosen). We accept J. R. Smith's evaluation[11,12,13] of the manganese bath measurements of ^{252}Cf nubar total, and we accept Boldeman's[14] evaluation of ^{252}Cf prompt nubar and nubar ratio measurements.

New measurements and analyses, made since the previous efforts, result in a more consistent fit to the data than were obtained in past efforts.

Recent Data and Analyses

Since Lemmel's paper[4] at the Washington Conference in 1975, there have been significant changes in some auxiliary data used to determine the parameters. The uranium half-lives have been evaluated here at NNDC[15] and the ^{234}U value has increased by about 0.4% compared to the most accurate values quoted in the literature. The latest experimental data on the half-life of ^{239}Pu have cleared up the discrepancy between calorimetric and alpha-counting half-lives and resulted in a decrease of 1.2%. These changes produced corresponding changes in the cross sections for the ^{235}U and ^{239}Pu whenever the amount of fissile material was determined by alpha counting, and this affected some of the most precise measurements in the input data set. Monte Carlo studies have refined the interpretations of certain measurements of alpha[16] (neutron capture to neutron fission cross-section ratio) and eta[17,18] (number of neutrons released per neutron absorbed).

However, the most extensive evaluation work has dealt with the determination of nubar. ^{252}Cf is used as a standard and the various fissile nuclide nubar values are measured as ratios to ^{252}Cf. Boldeman[14] and Smith[11,12,13] have evaluated various ^{252}Cf nubar measurements. Boldeman[19] has estimated a thickness correction for the foil samples in his earlier experiments on nubar ratios of the fissile nuclides to ^{252}Cf.

Following Smith's last review[13] in 1980, there has been little activity on the ^{252}Cf nubar problem although the results from liquid scintillator measurements tend to disagree with the results for manganese bath experiments. However, recently Axton[20] has commented on the Smith evaluation of Axton's experiment and Smith has replied.[21] Spencer[22] has published final results on his liquid scintillator measurement at Oak Ridge, and Edwards[23] has reported on a new measurement at Harwell.

Treatment of Uncertainty (Errors)

In general, the measurer's estimates of the uncertainties in their results are used - although we do not hesitate to follow evaluators' recommendations when they have shown good reason to change the original. We are indebted particularly to H.D. Lemmel and to B. R. Leonard, Jr. for their labors in this regard.

The LSF calculation scheme is such that the uncertainties (standard deviations) calculated for the output values are realistic if the uncertainties assigned to the input values are realistic. That is, if each experimental input datum is drawn from a population of data whose standard deviation is known

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to be equal to the experimental error assigned to it, then the LSF output errors are correct. We assume this contrary-to-fact situation to be the case. Even though there are good reasons for questioning the validity of this assumption, our results suggest that experimenters on the whole are realistic, even pessimistic, in assigning errors to their results. We find that not one of the individual data used differs from the LSF output value by more than twice the standard deviation of the difference between the two. The input data as a whole tend to be slightly more consistent with each other, judged by the Chi-squared test, than would be expected.

Results

The results of our fits for ^{233}U, ^{235}U, ^{239}Pu and ^{241}Pu are presented in Tables 1, 2, 3, and 4. The standards used in this work are shown in Table 5.

Table 1
^{233}U Results at 2200 m/s

Quantity	Lemmel(75)	Steen(72)	ENDF/B-V(79)	NNDC(8/82)
σ_{abs}	575.2±1.3	571.0±2.5	574.2	578.1±1.3
g_a	1.001±0.002	0.9990	0.9990	0.999±0.001
σ_{fiss}	529.9±1.4	525.1±2.4	528.4	530.2±1.2
g_f	0.9967±0.002	0.9968	0.9966	0.998±0.001
σ_γ	45.3±0.9	45.9±0.2	46.8	45.9±0.6
η	2.283±0.006	2.297±0.007	2.296	2.293±0.004
α	0.086±0.002	0.0874±0.0005	0.0868	0.0868±0.0011
ν_T	2.479±0.006	2.498±0.008	2.493	2.492±0.004
$^{252}Cf\ \nu_T$	3.746±0.009	3.783±0.014	3.766	3.767±0.004
$T_{1/2}(^{233}U)$	159000±200	159300±2300		159100±200

In Table 1, column 2 lists Lemmel's recommended values,[4] Column 3 lists Steen's evaluation[24], Column 4 lists the ENDF/B-V values, and Column 5 our most recent result.

Although Lemmel had later presented a talk at the 1977 Standards Meeting[5] at NBS, his conclusion was that the discrepancy between the fit to the 2200 m/s data and the fit to the 20°C Maxwellian data would not allow him to recommend either result. Accordingly, we have used his 1975 recommendations[4] in Column 2 to provide some comparison with our fit.

Our nubar and eta values are larger than Lemmel's due to our 0.6% larger value for ^{252}Cf nubar, which is primarily due to Spencer's measurement[22] performed after Lemmel's evaluation.

Table 2
^{235}U Results at 2200 m/s

Quantity	Lemmel(75)	Leonard(78)	ENDF/B-V(79)	NNDC(8/82)
σ_{abs}	680.9±1.7	681.9±1.8	681.9	681.5±1.2
g_a	0.980±0.003	0.9782	0.9781	0.9781±0.0009
σ_{fiss}	583.5±1.3	583.5±1.7	583.5	582.9±1.1
g_f	0.9758±0.002	0.9775±0.0011	0.9775	0.9771±0.0010
σ_γ	97.4±1.6	98.38±0.78	98.38	98.7±0.9
η	2.071±0.006	2.071±0.003	2.065	2.078±0.003
α	0.167±0.003	0.1686±0.0014	0.1686	0.1692±0.0017
ν_T	2.416±0.005	2.4205±0.012	2.437	2.430±0.004
$^{252}Cf\ \nu_T$	3.746±0.009		3.766	3.767±0.004
$T_{1/2}(^{234}U)$	244700±200			245700±500

In Table 2, we have listed Leonard's evaluation in Column 3 and the other Columns remain the same. Our ^{235}U nubar value is larger than the corresponding result of Lemmel by 0.6% due primarily to our higher ^{252}Cf nubar value. The increased half-life for ^{234}U reduces the ^{235}U fission cross section in Column 5 of Table 2. The ENDF/B fission cross-section value is based on Leonard's evaluation and as such is not an independent evaluation.

Beer's analysis[16] of Lounsbury's Chalk River alpha measurements results in a lower value for alpha (Maxwellian). However, Lemmel's g-factors for capture and alpha are much larger than ours and results in his larger Maxwellian alpha being reduced to a lower 2200 m/s alpha value compared to us.

Table 3
^{239}Pu Results at 2200 m/s

Quantity	Lemmel(75)	Leonard(81)	ENDF/B-V(79)	NNDC(8/82)
σ_{abs}	1011.2±4.1	1028.6±5.1	1011.9	1016.3±3.0
g_a	1.081±0.004	1.0782	1.0784	1.078±0.003
σ_{fiss}	744.0±2.5	754.8±4.5	741.7	748.3±1.7
g_f	1.0555±0.0024	1.0535±0.0015	1.0502	1.054±0.001
σ_γ	267.2±3.3	273.75±2.7	270.2	268.0±2.5
η	2.106±0.007	2.111±0.008	2.119	2.121±0.006
α	0.359±0.005	0.363±0.004	0.3643	0.358±0.003
ν_T	2.862±0.008	2.677±0.013	2.881	2.881±0.006
$^{252}Cf\ \nu_T$	3.746±0.009		3.766	3.767±0.004
$T_{1/2}(^{239}Pu)$	24290±70	24110		24100±12

Table 3 lists the results for ^{239}Pu. Once again, the half-life change has directly affected the fission cross section. Our value is 0.6% larger than Lemmel's value and 0.9% larger than the ENDF/B fission cross section. Our nubar value is again larger than Lemmel's and is due almost entirely to the increase in the ^{252}Cf standard. It can be noted that our larger nubar and fission cross section values produce an eta value which is 0.7% larger than Lemmel's value.

In the case of ^{239}Pu, the most recent shape measurements by Deruytter[25] (g_f = 1.0553 ± 0.0013) and Gwin[26] (g_f = 1.055 ± 0.002) agree better with Lemmel than with Leonard or ourselves, but the differences are minor. The most precise fission cross section measurement in our input set is also by Deruytter.[25] The half-life correction adjusts his reported value of 741.9 barns to an input value of 751.6 barns. Since his measurement carries a considerable weight in this fit, our fission cross section of 748.3 barns is significantly larger than both Lemmel's value and the ENDF/B-V value. Since Leonard's evaluation was performed much later than Lemmel's or the ENDF/B-V evaluation, he had the lower half-life value available and obtained a larger cross section as a result.

Table 4
^{241}Pu Results at 2200 m/s

Quantity	Lemmel(75)	Leonard(81)	ENDF/B-V(78)	NNDC(8/82)
σ_{abs}	1378.±9.	1388.5	1376.4	1373.7±10.5
g_a	1.039±0.003		1.043	1.044±0.002
σ_{fiss}	1015.±7.	1003.8	1015.0	1011.6±8.1
g_f	1.044±0.005		1.0452	1.046±0.005
σ_γ	362.±6.	364.7	361.4	362.1±5.4
η	2.155±0.010	2.160	2.178	2.167±0.007
α	0.357±0.007	0.363	0.3560	0.358±0.006
ν_T	2.924±0.010	2.953	2.953	2.943±0.009
$^{252}Cf\ \nu_T$	3.746±0.009		3.766	3.767±0.004

Most of our effort to date has dealt with the ^{235}U and ^{239}Pu data. However in order to complete the picture, we provide Table 4 which lists the present results for ^{241}Pu.

In Table 4, the various results listed for ^{241}Pu are generally consistent. Our eta value is 0.6% larger than Lemmel's value due entirely to the change in the ^{252}Cf nubar value.

In general, for all four fissile materials, we find no significant discrepancies between 2200 m/s and Maxwellian data. Like Lemmel, we have compared the fits obtained separately for each set of data. They are different, it is true; but the differences are neither large nor systematic. In neither set was the fitted value of any quantity different by more than twice its error from the value fitting the combined input data set.

We suggest that the recent evaluations and the different g-factors which we use have affected the Maxwellian data such that Lemmel's conclusion that these differ significantly from 2200 m/s data is no longer valid.

Table 5
Standards

Quantity	Value	Unit	Reference
$T_{1/2}(^{233}U)$	1.592±0.002	10^5 years	BNL-NCS-51320
$T_{1/2}(^{234}U)$	2.454±0.006	10^5 years	BNL-NCS-51320
$\sigma_\gamma(^{197}Au)$	98.65±0.09	barns	BNL-NCS-51388
$\sigma_\gamma(^{59}Co)$	37.18±0.06	barns	BNL-NCS-51388
$\sigma_\gamma(^{55}Mn)$	13.3±0.2	barns	BNL-NCS-51388
$\sigma_\alpha(^{10}B)$	3830.±6.	barns	BNL-NCS-51388
$\sigma_\alpha(^{6}Li)$	941 ±3.	barns	BNL-NCS-51388
g-factors	ENDF/B-V, Leonard		EPRI NP-167, EPRI NP-1763
g-error	Westcott, Leonard		AECL-3255, EPRI NP-167, -1763

Conclusion

Although we have not yet examined all the input data and revised them with the latest values of the auxiliary data, we have reevaluated those data sets which carry the largest weight in the fit, e.g. Deruytter's ^{235}U and ^{239}Pu fission cross section measurements,[25,27] the Rumanian ^{235}U and ^{239}Pu fission measurments,[28,29] Okazaki's alpha measurement,[30,31] and Bigham's and Keith's fission ratio measurements.[32,33,34]

We presented a preliminary version of this work at a BNL Conference.[34] We intend to complete the evaluation of the older (lower weight) measurements but we expect these changes to have a minor affect on the final recommendations. Contrary to Lemmel's 1977 paper,[5] we believe that the present situation is satisfactory in regard to choosing a best-fitting set of values for the 2200 m/s parameters of the fissile materials.

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