EPJ Web of Conferences **146**, 02021 (2017) DOI: 10.1051/epjconf/201714602021
ND2016

Evaluation of the ^{235}U resonance parameters to fit the standard recommended values

Luiz Leal[1,a], Gilles Noguere[2], Carlos Paradela[3], Ignacio Durán[4], Laurent Tassan-Got[5], Yaron Danon[6], and Marian Jandel[7]

[1] Institut de Radioprotection et de Sûrete Nucléaire, PSN-EXP/SNC/LNR, Fontenay-aux-Roses, France
[2] CEA, DEN, Cadarache, Saint-Paul-les-Durance, France
[3] Institute for Reference Materials and Methods Joint Research Center - European Commission, Geel, Belgium
[4] Universidade de Santiago de Compostela, Spain
[5] Centre National de la Recherche Scientifique/IN2P3-IPN, Orsay, France
[6] Rensselaer Polytechnic Institute, New York, USA
[7] Los Alamos National Laboratory, New Mexico, USA

Abstract. A great deal of effort has been dedicated to the revision of the standard values in connection with the neutron interaction for some actinides. While standard data compilation are available for decades nuclear data evaluations included in existing nuclear data libraries (ENDF, JEFF, JENDL, etc.) do not follow the standard recommended values. Indeed, the majority of evaluations for major actinides do not conform to the standards whatsoever. In particular, for the n + ^{235}U interaction the only value in agreement with the standard is the thermal fission cross section. A resonance re-evaluation of the n + ^{235}U interaction has been performed to address the issues regarding standard values in the energy range from 10^{-5} eV to 2250 eV. Recently, ^{235}U fission cross-section measurements have been performed at the CERN Neutron Time-of-Flight facility (TOF), known as n_TOF, in the energy range from 0.7 eV to 10 keV. The data were normalized according to the recommended standard of the fission integral in the energy range 7.8 eV to 11 eV. As a result, the n_TOF averaged fission cross sections above 100 eV are in good agreement with the standard recommended values. The n_TOF data were included in the ^{235}U resonance analysis that was performed with the code SAMMY. In addition to the average standard values related to the fission cross section, standard thermal values for fission, capture, and elastic cross sections were also included in the evaluation. This paper presents the procedure used for re-evaluating the ^{235}U resonance parameters including the recommended standard values as well as new cross section measurements.

1. Introduction

Prior to describing the issues with regard to the standards for ^{235}U it is worthwhile to present a succinct description of the ^{235}U evaluation work carried out over the last decades. In the later 1980s and early 1990s a ^{235}U Reich-Moore resonance evaluation was performed from thermal to 2.25 keV [1] using the code SAMMY [2]. It was the first attempt to use a more rigorous resonance formalism to address issues with interference effects in the fission channels. The evaluation represented a huge improvement compared to previous ^{235}U evaluations for which the Single-Level Breit-Wigner (SLBW) formalism was used together with background cross sections to make up for the SLBW deficiency to represent fissile isotope. However, very little integral benchmark testing were carried out to assess the evaluation effectiveness prior its inclusion in the evaluated files. The evaluation was adopted in evaluated nuclear data projects and then underwent series of benchmark testing. The testing included sensitivity analysis and cross section adjustments based on benchmark experiments. The results demonstrated that the evaluation performed poorly mainly due to concerns with the capture cross-section in the energy region 22.6 eV to 454 eV [3] indicating a need for increasing the capture cross section. However no issue with the fission cross-section was found. A close inspection of the problem revealed that a low value of the average gamma-capture width was responsible for the very low capture cross section. It should be pointed out that no reliable capture cross section measurements existed at the time in the energy range above 100 eV. There existed capture cross-section data but these data were not systematically included in the evaluation due to issues such as normalization and background. Hence the ^{235}U evaluation was revised on the basis of integral results and sensitivity analysis. The revised ^{235}U evaluation was made available and included in the ENDF, JEFF and JENDL libraries [4]. The JENDL project adopted the evaluation up to 500 eV and used an unresolved resonance representation above 500 eV to help improving the results of fast critical assembly benchmark (FCA) [5]. The revised evaluation gave a high capture cross-section that did not support the FCA benchmark results. A similar scenario observed with the FCA benchmark results were also indicated with the ICSBEP HEU-MET-INTER-006 benchmark series (ZEUS benchmarks) [6]. These divergences with integral

---

[a] e-mail: `luiz.leal@irsn.fr`

DOI: 10.1051/epjconf/201714602021

benchmark calculations prompted the proposal for a subgroup of the Working Party on International Nuclear Data (WPEC) to investigate the ^{235}U capture problem [7]. The results of the WPEC criticality calculations showed an overestimation of the ^{235}U capture cross-section of about 10%. The WPEC recommendation was that rather than re-evaluating the ^{235}U resonance parameters based solely on integral benchmark results new capture cross-section measurements should be made to confirm the findings of the WPEC subgroup. Hence, TOF capture cross-section measurements were planned and performed independently at the Rensselaer Polytechnic Institute (RPI) [8] and at the Los Alamos National Laboratory (LANL) [9]. These new measurements were used together with the computer code SAMMY to update the ^{235}U resonance parameters in the energy range from thermal to 2.25 keV. The results demonstrated an improvement in benchmark results. However, despite all efforts to address the capture issue in the resonance region the problem with the standard cross section still remained. Recently measured fission cross-section data carried out at the n_TOF facility provided strong support to the fission standard values. Indeed, the normalization of the n_TOF fission data in the energy range 7.8 eV to 11.0 eV supported the standard value in this energy range but also reinforced the standard averaged fission cross-section values [10] in the resonance region. Hence, the resonance parameter evaluation was revised with the inclusion of the n_TOF experimental fission data.

2. ^{235}U resonance evaluation in the energy range 10^{-5} eV to 2.25 keV

In this section a description of the ^{235}U evaluation using the computer code SAMMY is presented including resonance analysis and external level determination. The experimental data base used in the evaluation is described with emphasis on new measurements added to the present evaluation.

2.1. External energy levels determination

Previous evaluations of the ^{235}U resonance parameters made use of several external resonance energies, namely 14 bound levels and 14 levels above 2.25 keV, which proved to be unnecessary to represent the interference effects in the resonance range 10^{-5} eV to 2.25 keV. Actually, it was apparent that the issue in connection with the fitting of the standard fission cross section was intimately related to the external energy levels. The long-range interference effects inherent in the R-matrix methodology precluded finding a good fitting of the experimental fission data. It also had an impact on the elastic scattering cross section. The present evaluation contains five bound energy levels and five energy levels above 2.25 keV. Included in the bound levels there is a negative energy close to zero with a very tiny neutron width which is responsible for the bending of the energy dependence of the η(E) at low energy [11]. The 10 external energy levels are listed in Table 1 in which for each the resonance energy E_r, gamma width Γ_γ, neutron width Γ_n, two fission widths Γ_{f1} and Γ_{f2} and the spin and parity J^π are shown. Negative signs associated with the fission partial widths Γ_{f1} and Γ_{f2} reflect the positive-negative sign of the reduced amplitude width γ_{f1} and γ_{f2}. It follows a convention established in the ENDF resonance parameters representation. The effect of the external level is shown in Fig. 1 which corresponds to the scattering cross section in the energy range 10^{-5} eV to 2.25 keV without the presence of resonances. In the plateau around the energy of 1125 eV the scattering cross-section due to the external energy values is about 11.194 barns corresponding to a scattering radius of 9.438 fm. An accurate representation of the external resonance contribution provides the grounds to determining the effective scattering radius. The analysis of high-resolution transmission data led to an effective scattering radius of 9.602 fm.

Table 1. Energy bound levels and energies above 2.25 keV.

E_r (eV)	Γ_γ (meV)	Γ_n (meV)	Γ_{f1} (meV)	Γ_{f2} (meV)	J^π
Energy bound Levels					
−75.405	47.781	507.274	−487.090	−443.345	3^-
−5.253	36.797	12.170	195.681	−160.038	4^-
−0.481	39.228	0.088	129.661	−80.535	3^-
−0.432	38.024	0.033	167.072	−8.283	4^-
-3.657×10^{-5}	39.988	6.461×10^{-8}	−0.509	0.935	4^-
Energy levels above 2.25 keV					
2281.325	44.083	12.459	155.711	458.850	4^-
2284.014	41.147	3802.461	1956.501	22.864	3^-
3312.563	47.228	11457.530	474.421	571.292	3^-
3819.129	38.494	1242.316	−511.662	67.709	4^-
4500.997	33.681	33.8.548	286.623	364.141	3^-

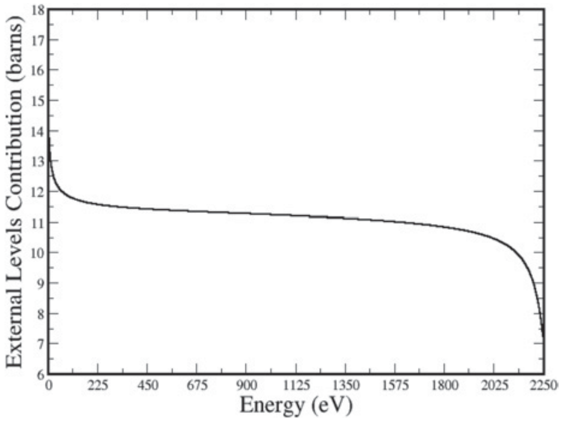

Figure 1. External levels contribution on the scattering cross-section in the energy ranges 10^{-5} eV to 2.25 keV.

2.2. Experimental data base

The novel feature of the work described in this paper, in contrast to previous work on the ^{235}U resonance parameter evaluation, is the addition of new cross-section measurements done at LANL, RPI and at n_TOF. Capture cross section measurements done at LANL and RPI were central to unveil issues with the capture cross section above 100 eV. The fission cross-section measurements carried out at n_TOF supported the standard cross section values. Furthermore, the evaluation was done including high-resolution transmission, fission cross-section, and eta measurements that were accounted for in previous ^{235}U evaluations. The experimental data used in the resonance parameter evaluation are displayed in Table 2 in which

Table 2. Experimental Data Included in the SAMMY Resonance Analysis.

Reference	Energy Range (eV)	Data
Transmission		
Spencer (ORNL/1984) [12]	0.01–8.0	L = 18 meters, n = 0.001468 atom/barn, and T = 293.3 K
Harvey (ORNL/1986) [13]	0.4–68.0	L = 18 meters, n = 0. 03269 atom/barn, and T = 77 K
Harvey (ORNL/1986) [13]	4.0–2250.0	L = 80 meters, n = 0. 00233 atom/barn, and T = 77 K
Harvey (ORNL/1986) [13]	4.0–2250.0	L = 80 meters, n = 0. 03269 atom/barn, and T = 77 K
Fission		
Gwin (ORNL/1984) [14]	0.1–20.0	L = 25.6 meters and T = 293.6 K
Weston (ORNL/1992) [15]	100.0–2250.0	L = 86.5 meters and T = 293.6 K
Weston (ORNL/1984) [16]	14.0–2250.0	L = 18.9 meters and T = 293.6 K
Paradela (n_TOF/2010) [17]	0.7–2250.0	L = 185 meters and T = 293.6 K
Danon (RPI/2011) [8]	100.0–2250.0	Yield L = 25.56 meters and T = 293.6 K
Wagemans [18]	0.001–0.4	L = 18 meters and T = 293.6 K
Eta(η)		
Wartena (Geel/1987) [19]	0.0018–1.0	L = 8 meters and T = 293.6 K
Weigmann (ILL/1990) [20]	0.0015–0.15	Chopper, T = 293.6 K
Capture		
Danon (RPI/2011) [8]	100.0–2250.0	Yield L = 25.56 meters and T = 293.6 K
Jandel (LANL/2012) [9]	100.0–2250.0	L = 25.45 meters and T = 293.6 K
Perez (ORNL/1973) [21]	0.01–200.0	L = 39.7 meters and T = 293.6 K
De Saussure (RPI/1967) [22]	0.01–2250.0	L = 25.2 meters and T = 293.6 K

TOF length is indicated by the letter L, thickness by n, and temperature by T. The Reich-Moore approach of the code SAMMY was used for fitting the data. A total of 3170 resonance levels were identified in the energy range 0–2250 eV to reproduce the experimental data.

2.3. ^{235}U Resonance parameter evaluation

Before starting the fitting of the data shown on Table 2 a careful examination of the experimental conditions was done. Experimental resolution, normalization, background, multiple-scattering, data alignment, etc., were inspected to assure consistency with the data set. A sequential analysis of the data shown in Table 2 was carried out with the code SAMMY to achieve a reasonable fit of the data with an acceptable χ^2. Not only the resonance parameters were let to vary but also normalization, resolution parameters, etc. were also searched.

The two experimental η(E) values at the low energy were fitted with the SAMMY code. The η(E) shape observed in the experimental is followed by the fitting of the data. The bending effect perceived is the result of the interference effects of the bound level placed at the energy -3.657×10^{-5} eV with a very tiny neutron width. The fitting is displayed in Fig. 2.

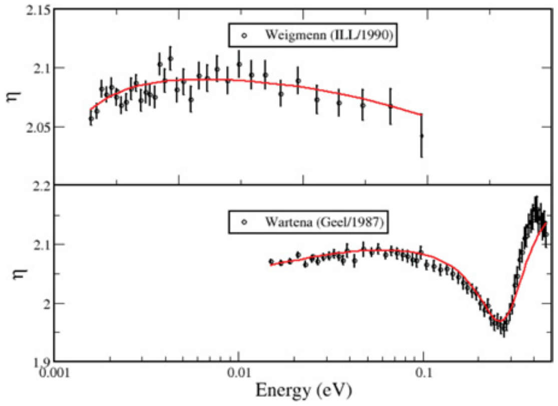

Figure 2. Comparison of the experimental and calculated η(E) in the thermal energy range.

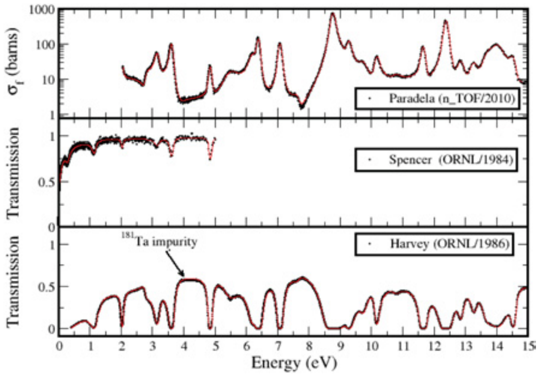

Figure 3. Comparison of the SAMMY fit of the experimental data.

The n_TOF fission data were normalized based on the fission integral in the energy range 7.8 eV to 11 eV. A decision was made to not use the normalized data but rather to include the fission integral value in the fitting process. It was noted that, as a result of this procedure, the average values of the fission cross section related to the standard were straightforwardly fitted. The results of the fitting of the Harvey [13], Spencer [12], and the Paradela [17] are shown in Fig. 3. An examination of the transmission data of Harvey revealed an inconsistency with the remaining data set around the energy 4.25 eV. It was discovered that the issue was due to an impurity of ^{181}Ta present in the target sample. Although the data reduction was suitably done it appears that the ^{181}Ta impurity was not completely removed.

It is interesting to note that the fission integral value in the energy region 7.8 eV to 11.0 eV led to a normalization of the Weston [15, 16] and Gwin [14] fission data of about 2%. The fitting of the Weston [15] and Paradela [17] is shown in Fig. 4 in the energy range 100 eV to 400 eV. The resolution of the n_TOF data is excellent displaying the details of the Porter-Thomas like fluctuations not seeing in the Weston data in this energy range.

Two measured capture data at LANL and RPI were used in the resonance evaluation above 100 eV. The capture

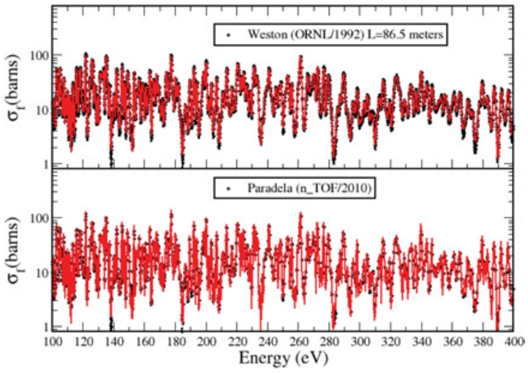

Figure 4. SAMMY fitting of the fission cross section in the 100 eV to 400 eV energy range.

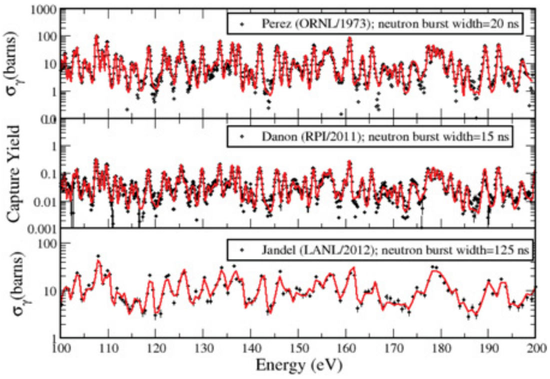

Figure 5. SAMMY fitting of the fission cross section in the 100 eV to 200 eV energy range.

data of Perez [21] were used below 200 eV. The three data sets are displayed in Fig. 5 together with the SAMMY fit in the energy range 100 eV to 200 eV. As can be seen the resolution of the RPI data is excellent for the use in the resonance analysis. The time-of-flight lengths for the three measurements are about the same with the main difference on the neutron burst width. In the energy range 100 eV to 200 eV the SAMMY fit was based on the Perez, Danon, and Jandel capture data. Above 200 eV the fitting relied mainly on the RPI data.

2.4. Thermal values, average values and the standard data

As previously indicated the main motivation for revising the ^{235}U resonance parameter evaluation is to address issues with the quoted standard values [10]. The two major changes in the present evaluation relative to the thermal cross section standard (energy of 0.0253 eV) are the capture and scattering thermal cross section values. Previous ^{235}U resonance parameters evaluation replicates the thermal fission cross-section standard values. The revision of the external resonance values, bound energy levels and energies above 2250 eV, allowed a quick convergence to the standard thermal values and the average values for the fission cross section. The thermal values calculated with SAMMY and the new resonance parameters are shown in Table 3. It is also listed the fission integral in the energy range 7.8 eV to 11 eV.

Table 3. Standard values and resonance parameters results for 0.0253 eV.

Parameter	Standard values (barns)	Values obtained with the new resonance parameter (barns)
σ_f (**barns**)	584.380 ± 1.030	584.417
σ_γ (**barns**)	99.304 ± 0.725	99.231
σ_s (**barns**)	14.087 ± 0.219	14.086
Fission integral in the 7.8 eV–11 eV range (**barns.eV**)	246.396 ± 1.244	246.854

Table 4. Standard average fission integral.

Energy Interval (eV)	Standard values (barns)	Average fission cross section obtained with the new resonance parameter (barns)
100–200	21.17 (11)*	21.02
200–300	20.69 (11)	20.77
300–400	13.13 (7)	13.22
400–500	13.78 (8)	13.49
500–600	15.17 (9)	15.20
600–700	11.51 (7)	11.53
700–800	11.10 (6)	11.10
800–900	8.21 (5)	8.15
900–1000	7.50 (4)	7.37
1000–2000	7.30 (4)	7.29

*Read as 21.17 ± 0.11.

The results presented in Table 3 are in agreement with the standard values. The fission cross section data measurements performed at CERN, Paradela [17], were fundamental in reproducing the standard values. Furthermore, the standard average fission integral from 100 eV up to 2000 eV were also replicated with calculations using the revised resonance parameters. The average fission cross section values compared with the standard are shown in Table 4.

3. Conclusions

Reevaluation of the ^{235}U resonance parameter was carried out with the code SAMMY to address issues with thermal cross section standards and average fission cross-section values. A new fission cross section measurement done at CERN was the chiefly factor for obtaining a good agreement between calculated and experimental fission values. The new set of resonance parameters include less external energy levels and provide a better way of calculating the interference effects on the fission channels. Thermal result values are within the indicated standard uncertainties. The fission integral in the energy range 7.8 eV to 11 eV is well reproduced. The normalization of the n_TOF fission data in the energy range 7.8 eV to 11.0 eV supported the standard values above 100 eV up to 2250 eV. The SAMMY fit of the data replicated the average values within the quoted uncertainties. The new set of resonance parameters presented in this paper has been greatly improved compared with previous versions.

References

[1] G. de Saussure, L.C. Leal, R. Perez, N.M. Larson, and M.S. Moore, Nucl. Sci. Eng. **103**, 109 (1989)

[2] N.M. Larson, *Updated Users' Guide for SAMMY: Multi-level R-Matrix Fits to Neutron Data Using Bayes's Equations*, ENDF-364/R2, Oak Ridge National Laboratory USA (2008). Available at Radiation Safety Information Computational Center (RSICC) as PSR-158

[3] Stéphane Cathalau, Amal Benslimane, Abdelmajid Maghnouj, and Philippe Fougeras,"Qualification of the JEFF2.2 Cross Sections in the Epithermal and Thermal Energy Ranges Using a Statistical Approach," Nucl. Sci. Eng. **121**, 326 (1995)

[4] L.C. Leal, H. Derrien, N.M. Larson, and R.Q. Wright, "R-Matrix Analysis of ^{235}U Neutron Transmission and Cross-Section Measurements in the 0- to 2.25-keV Energy Range," Nucl. Sci. Eng. **131**, 230 (1999)

[5] M. Fukushima, Y. Kitamura, T. Kugo and S. Okajima, "Benchmark models for criticalities of FCA-IX assemblies with systematically changed neutron spectra," J. Nucl. Sci. Technol., Published online: Jun (2015)

[6] International Handbook of Evaluated Criticality Safety Benchmark Experiments, NEA/NSC/DOC(95)03, OECD Nuclear Energy Agency September 2006 (rev)

[7] NEA/NSC/WPEC/DOC(2011)433, Final Report of WPEC Subgroup 29 on 235U Capture Cross-section in the keV to MeV Energy Region, Nuclear Energy Agency, Organization for Economic Co-operation and Development, 2011

[8] Y. Danon, "Nuclear Data Measurements at RPI," 2011 Cross Section Evaluation Working Group Meeting, National Nuclear Data Center, Brookhaven National Laboratory, Upton, NY, November 16, 2011

[9] M. Jandel et al., "New Precision Measurements of the ^{235}U(n,γ) Cross Section," Phys. Rev. Lett. **109** (2012)

[10] Carlson A.D. et al. "International Evaluation of Neutron Cross Section Standards," N. D. S. (2009) 3215-3324 (pg 3291). See also `https://www-nds.iaea.org/standards/Data/endf-6-format/std-092_U_235.endf`

[11] L. Erradi, A. Santamarina, and O. Litaize, "The Reactivity Temperature Coefficient Analysis in Light Water Moderated UO_2 and UO_2-PuO_2 Lattices," Nucl. Sci. Eng. **144**, 47 (2003)

[12] R.R. Spencer, J.A. Harvey, N.W. Hill, and L. Weston, Nucl. Sci. Eng. **96**, 318 (1987)

[13] J.A. Harvey, N.W. Hill, F.G. Perey, G.L. Tweed, and L.C. Leal, Proc. Int. Conf. on Nuclear Data for Science and Technology, Mito, Japan, May 30–June 3, 1988

[14] R. Gwin, R.R. Spencer, R.W. Ingle, J.H. Todd, and S.W. Scoles, Nucl. Sci. Eng. **88**, 37 (1984)

[15] L.W. Weston and J.H. Todd, Nucl. Sci. Eng. **111**, 415 (1992)

[16] L.W. Weston and J.H. Todd, Nucl. Sci. Eng. **88**, 567 (1984)

[17] C. Paradela et al. "High accuracy 235U(n,f) data in the resonance region," The European Physical Journal, Vol. 111 (2016), WONDER-2015 4th International Workshop on Nuclear Data Evaluation for Reactor Application, Aix-en-Provence, France, 5–8 October 2015

[18] C. Wagemans, P. Schillebeeckx, A.J. Deruyter, and R. "Subthermal Fission Cross Section Measurements for ^{235}U and ^{239}Pu," Nuclear Data for Science and Technology, p. 91, Mito, Japan (1988)

[19] J.A. Wartena, H. Weigmann, and C. Burkholz, Report IAEA Tecdoc **941**, 123 (1987)

[20] H. Weigmann, P. Geltenbort, B. Keck, K. Shrenckenbach, and J.A. Wertena, Proc. Intern. Conf. on The Physics of Reactors, Marseille **P1**, 133 (1990)

[21] R.B. Perez, G. de Saussure, and E.G. Silver, Nucl. Sci. Eng. **52**, 46 (1973)

[22] G. de Saussure, R. Gwin, L.W. Weston, and R.W. Ingle, "Simultaneous Measurements of the Neutron Fission and Capture Cross Section for 235U for Incident Neutron Energy from 0.4 eV to 3 keV," ORNL/TM-1804, Martin Marietta Energy Systems, Inc., Oak Ridge National Laboratory; Oak Ridge, TN (1967)