Nuclear Data Testing at AECL

Dan Roubtsov
AECL, Chalk River Laboratories
Canada

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Outline

- n +^{16}O at low neutron energy

- TSL (S(α,β)) for H_2O, D_2O

Collaboration with

K. Kozier (retired), J.C. Chow (AECL),

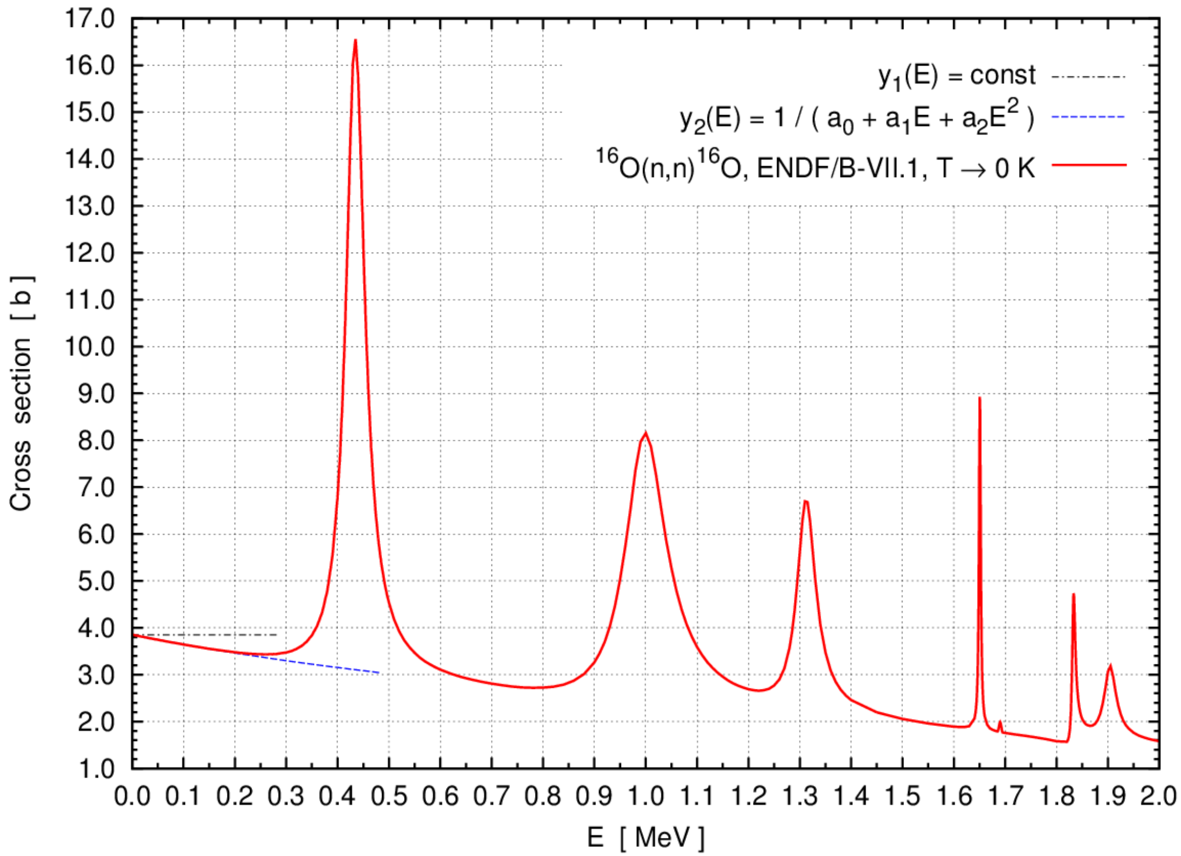
A. Plompen, S. Kopecky (IRMM, Belgium),

J.I. Márquez Dámian (Bariloche, Argentina),

J. Svenne (U of Manitoba, Canada)

L. Canton (Istituto Nazionale di Fisica Nucleare, Italy)

Elastic neutron scattering cross sections of ^{16}O at low *E* (from nuclear physics standpoint)

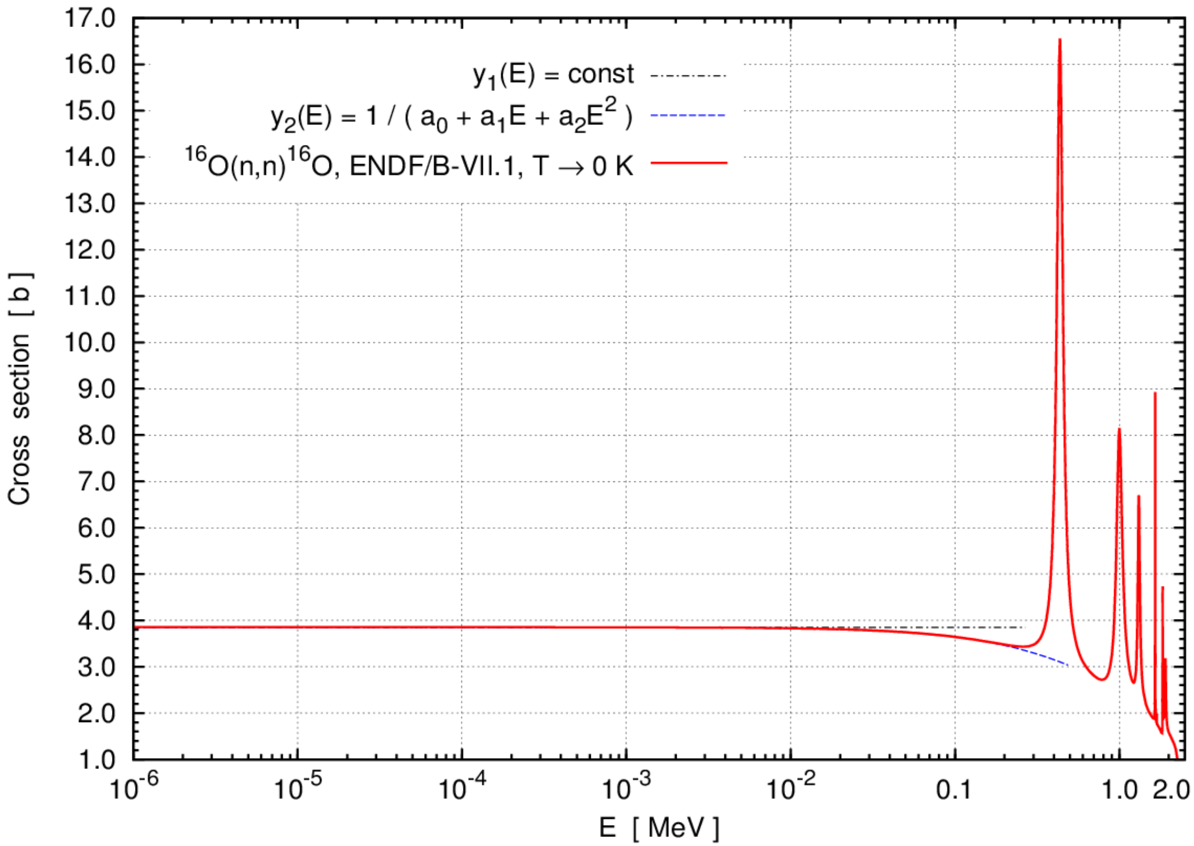


- In the limit of $E \to 0$ (better, ***E* << 0.1 MeV**),

  $\sigma_s(^{16}\mathrm{O})$ *vs.* *E* approaches a constant (≈ 3.85 b in ENDF/B-VII.1)

This *E* = 0 limit is for neutron scattering x-sections at **T = 0 K,**
or ***n* scattering on target-at-rest, free nuclide (MF3, MT2 in ENDF-6 file)**

Elastic neutron scattering cross sections of ^{16}O at low *E* (for Reactor Physics applications)



- At ***E*** **< 0.01 MeV**, σ_s**(E)** ≈ ***const*** works well, and *const* = $\sigma_{s,\,th}$.

For ***E*** **< 0.2 MeV,** $\sigma_s(E) \approx 1 / (a_0 + a_1 E + a_2 E^2)$, (effective-range approx.).

If **T ≠ 0 K, Doppler broadening** modifies $\sigma_s(E) \rightarrow \sigma_s(E;\ \mathbf{T})$ at ***E*** **< 1 - 10 eV; usually, use F**ree **G**as **M**odel, but also ^{16}O-in-UO_2, ^{16}O-in-D_2O, *etc.*

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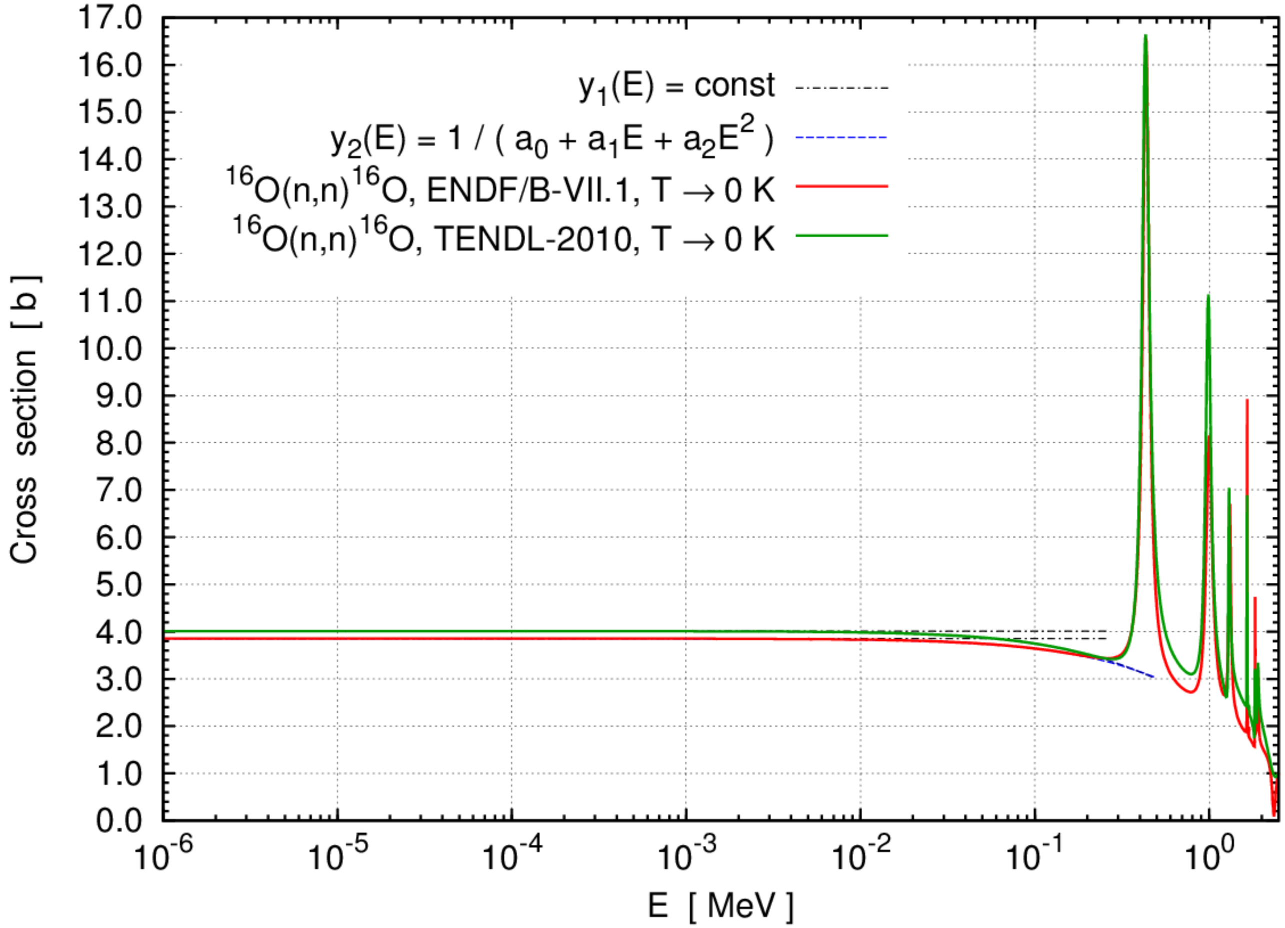
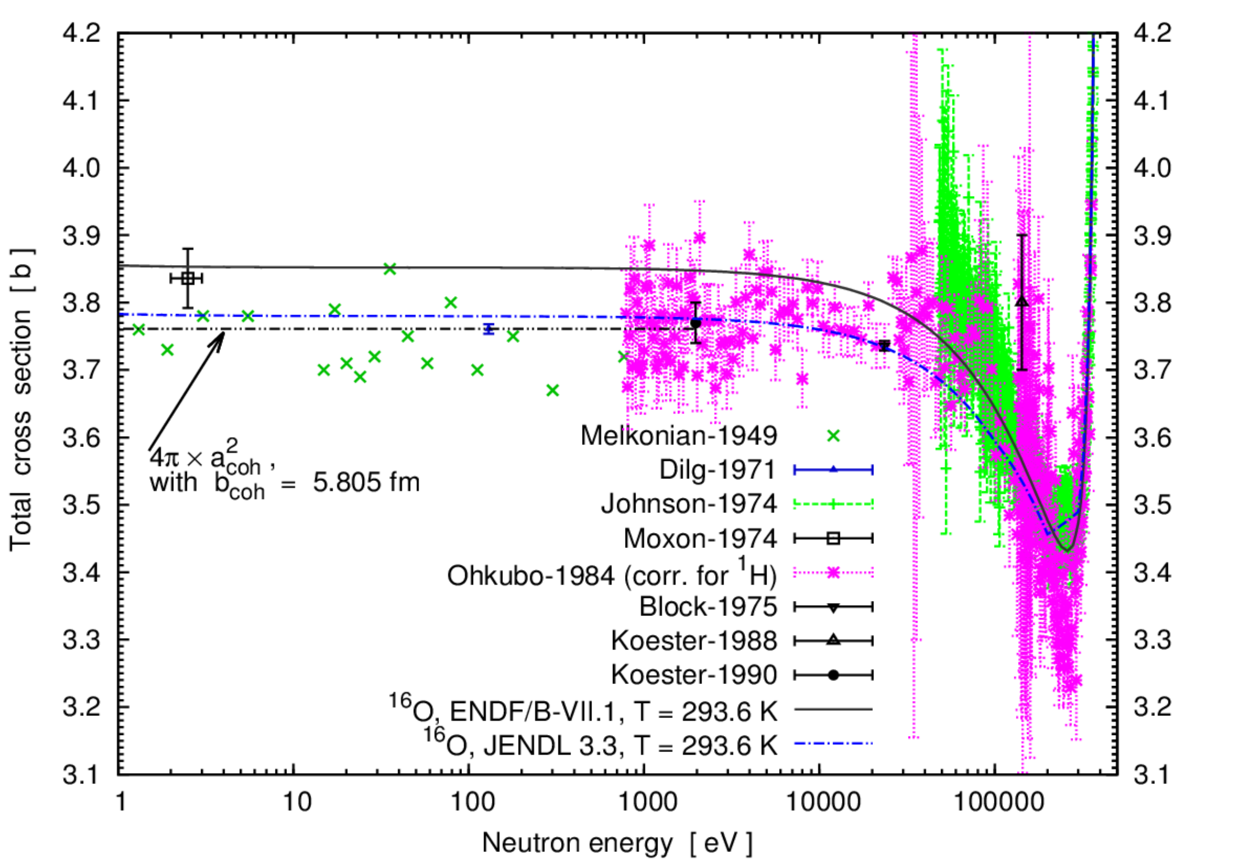
Elastic neutron scattering cross sections of ^{16}O at low *E* (for Reactor Physics applications)



- If **T** $\neq$ **0 K, Doppler broadening** modifies $\sigma_s(E) \rightarrow \sigma_s(E; \mathbf{T})$ at ***E* < 1 - 10 eV; usually, use Free Gas Model**, but need ^{16}O-in-UO_2, ^{16}O-in-BeO, ^{16}O-in-D_2O, *etc.*
- **TSL** needs $\sigma_{s,\,th} \pm \Delta\sigma_{s,\,th}$, so check $\sigma_{s,\,th}$ of ^{16}O in **different evaluations …**

Measurements at low energy (E < 0.3 – 0.4 MeV)



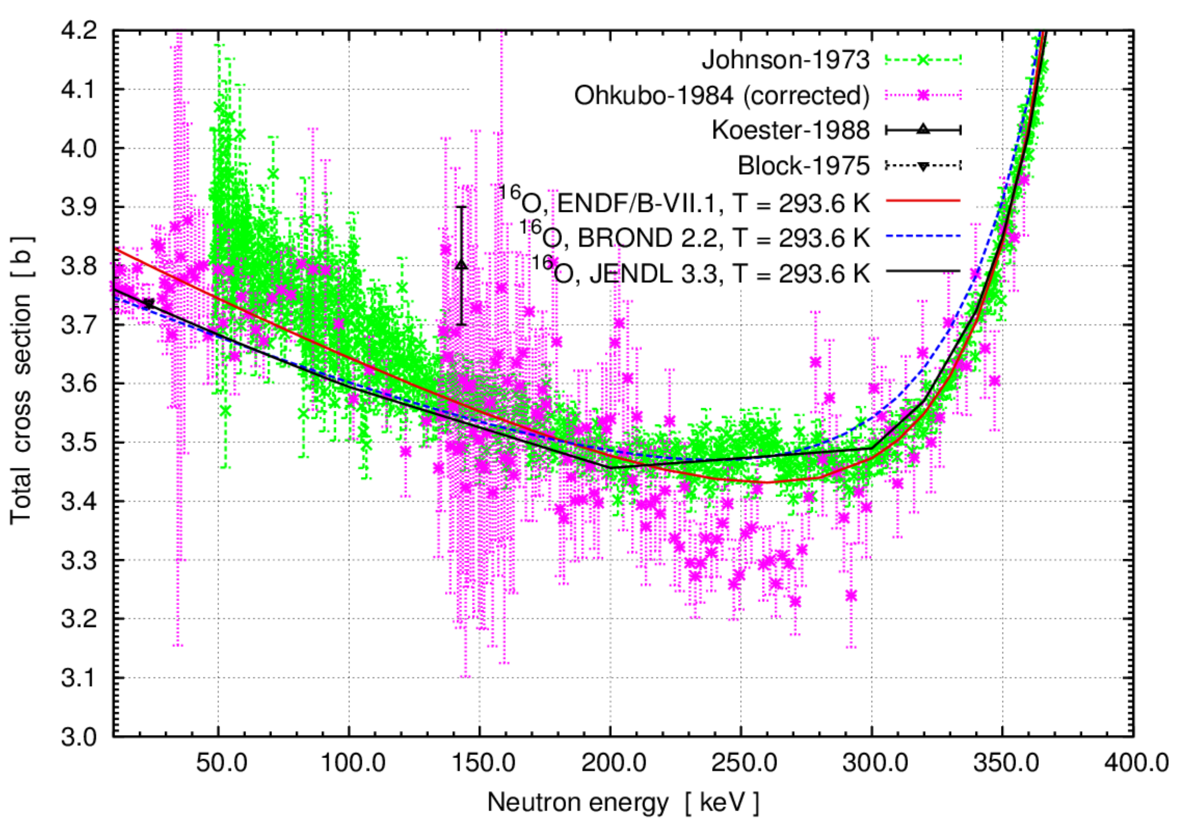
- **Johnson-1974**: unexpected behaviour at E < 0.2 MeV , σ_{tot} from **BeO** and **Be**, sample quality **?**

C.H. Johnson *et al*., "*The O-16 + n total cross section: diagnostics and refinements*," Report **ORNL-4937**, pp. 195-196 , (1974).

- **Ohkubo-1984**: σ_{tot} from **Al_2O_3** and **Al**; data corrected for **H** in Al_2O_3 (A. Plompen & S. Kopecky)

M. Ohkubo, "*Neutron Total Cross Section Measurements on Oxygen, Aluminum and Carbon below 930 keV*," Report **JAERI-M 86-193**, Japan Atomic Energy Research Institute (1987).

Measurements at low energy (*E* < 0.3 – 0.4 MeV)

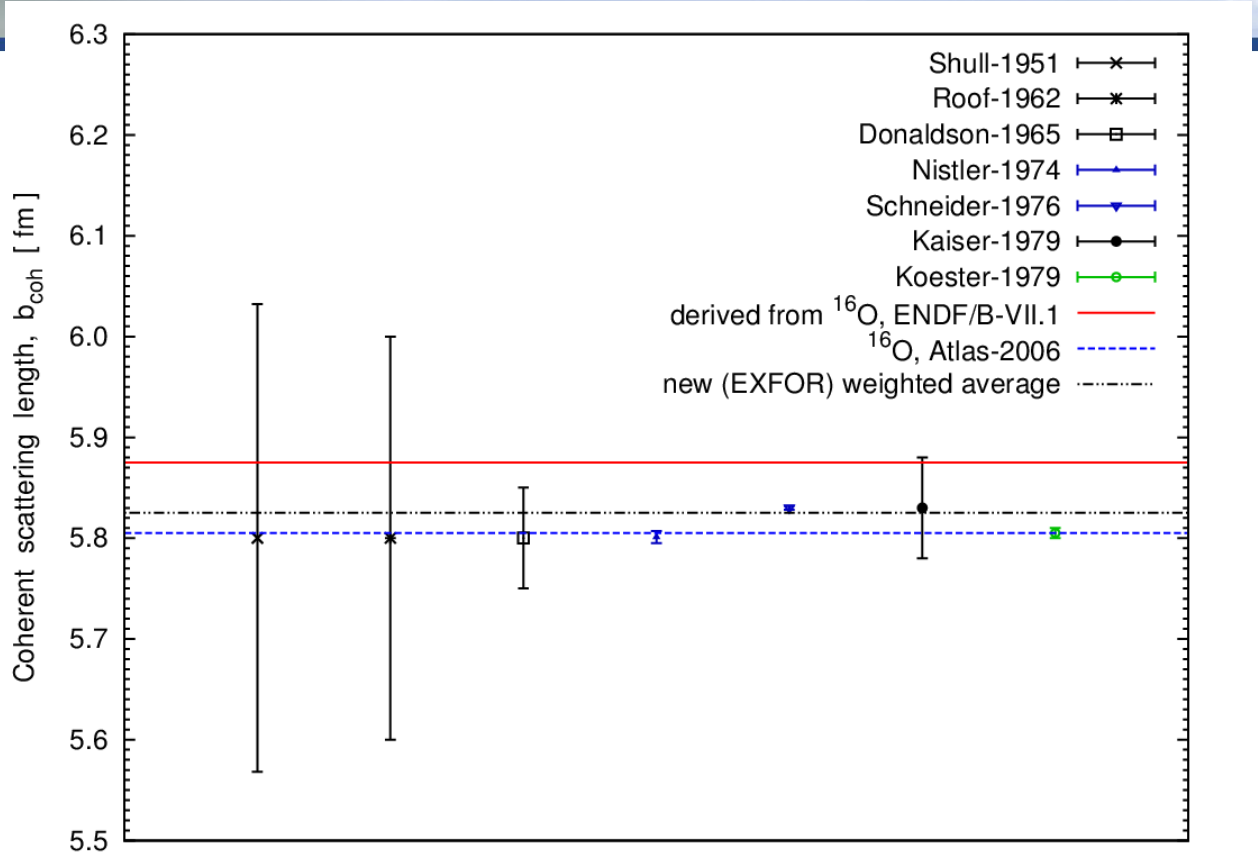


- **Total cross-sections near a minimum at $E \approx$ 250 keV (E ~ 100 - 300 keV):**

NEW evaluation(s): we can estimate $\sigma_{tot}(E) \pm \Delta\sigma_{tot}(E)$ (or $\sigma_s(E) \pm \Delta\sigma_s(E)$),
*but can we compare it with **accurate** measurements* **?**
(*e.g., we have **Block-1975**,* E = ***23.5 keV***, σ_{tot} = *3.736* $\pm$ *0.007 b* (**±0.19%**) , *what else ?*)

This could be a test for the accuracy of ***R′***(^{16}O) ~ 5 fm (σ_{pot}) and res. parameters,
$\mathbf{E_j}$, Γ_n, Γ_γ that define σ_{tot} at low energies ~ 10 keV < E < 300 keV :
local bound level(**s**), E_0 ~ **–3** MeV (*s-res.*, Γ_n ~ $|E_0|$) and $E_1 \approx$ 0.434 MeV (*p-res.*)

Neutron Optics: b_{coh} [fm] of ^{16}O ($J = 0$)



- **New weighted average from EXFOR experimental data:**
  b_{coh} = 5.824 ± 0.002 fm (A. Plompen & S. Kopecky)
- **Atlas-2006 estimate:** b_{coh} = 5.805 ± 0.005 fm
- Derived from ^{16}O, ENDF/B-VII.1: b_{coh} = 5.874 ± 0.059 fm

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b_{coh} [fm] and $\sigma_{s,\,th} = \sigma_{coh}$ [barn] of ^{16}O

- **Atlas-2006** ($\sigma_{s,\,th} \leftrightarrow b_{coh}$ consistent):

$\sigma_{s,\,th}(^{16}\mathrm{O}) = 3.761 \pm 0.006$ b (±**0.16%**)

$b_{coh}(^{16}\mathrm{O}) = 5.805 \pm 0.005$ fm (±0.09%)

- **ENDF/B-VII.1** :

$\sigma_{s,\,th}(^{16}\mathrm{O}) = 3.852 \pm 0.077 \text{ b} \rightarrow b_{coh}(^{16}\mathrm{O}) = 5.874 \pm 0.059 \text{ fm}$

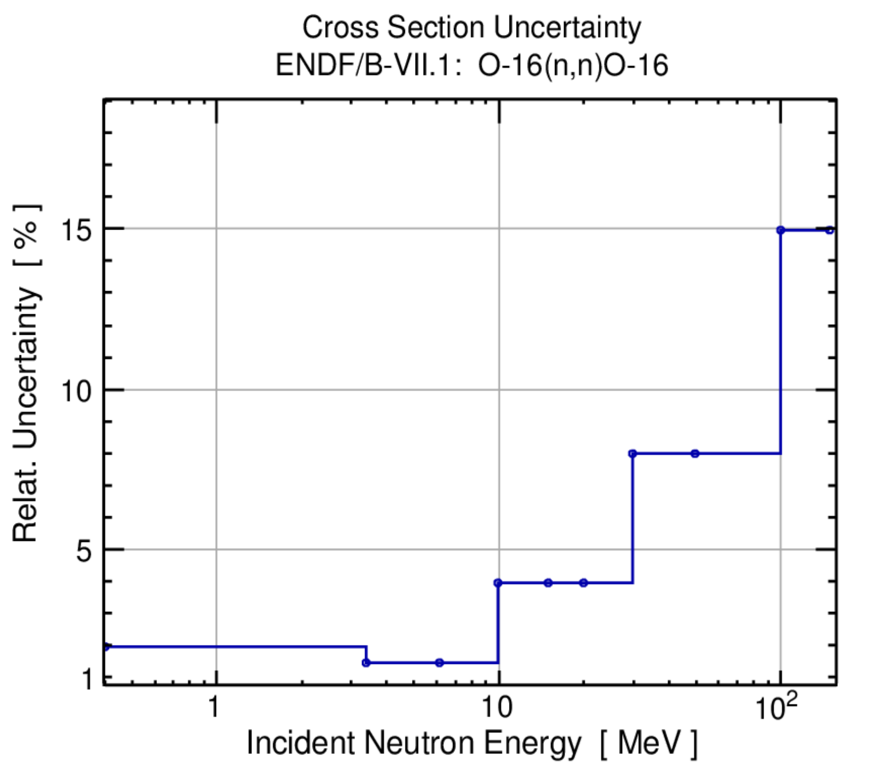
- **JENDL-4.0**:

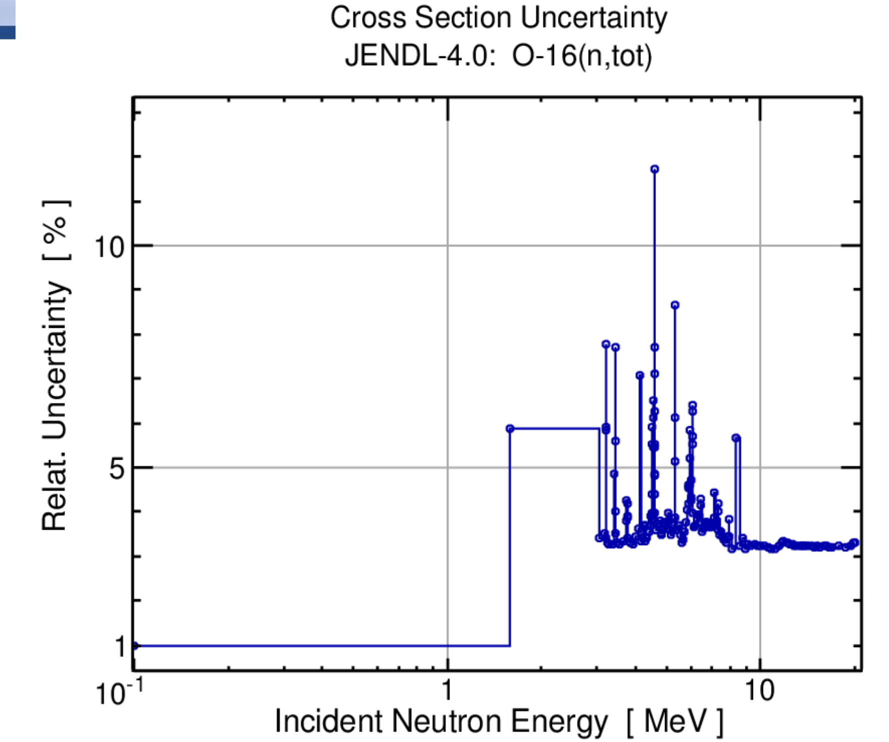
$\sigma_{s,\,th}(^{16}\mathrm{O}) = 3.841 \pm 0.038 \text{ b} \rightarrow b_{coh}(^{16}\mathrm{O}) = 5.866 \pm 0.029 \text{ fm}$

- **New (EXFOR-based) estimate**:

$\sigma_{s,\,th}(^{16}\mathrm{O}) = 3.786 \pm 0.003 \text{ b} \leftarrow b_{coh}(^{16}\mathrm{O}) = 5.824 \pm 0.002 \text{ fm}$

Uncertainty of n + ^{16}O at low neutron *E*





- Relative uncertainty of $\sigma_{tot}(E)$ *and* $\sigma_s(E)$ calculated using ^{16}O covariance file (MF=33)

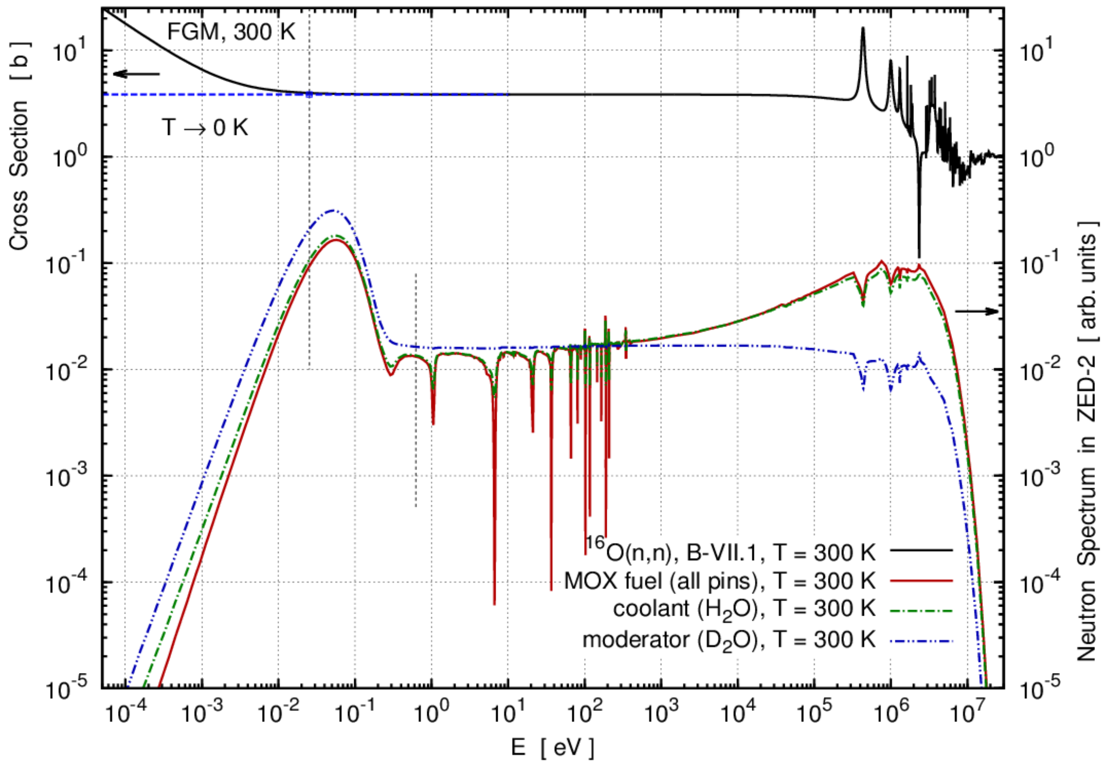
  ENDF/B-VII.1: $\Delta\sigma_{tot}/\sigma_{tot} \approx \Delta\sigma_s/\sigma_s$ = **± 2.0%** at *E* < **3.4 MeV**.

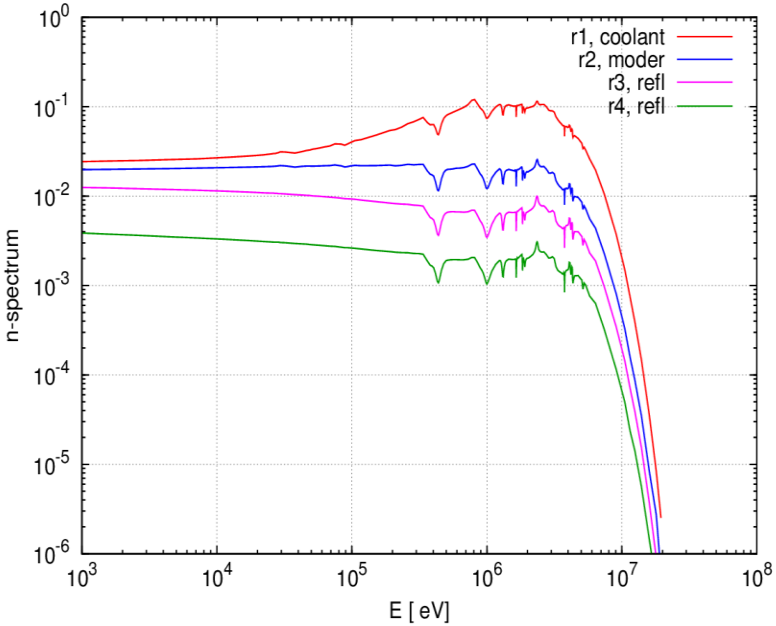
  MF=33, MT=2, $\mathbf{cov}_s(i, i')$: 10^{-5} eV < E_1 < 0.4 MeV, 0.4 MeV < E_2 < 3.4 MeV, ...

  JENDL-4.0: $\Delta\sigma_{tot}/\sigma_{tot} \approx \Delta\sigma_s/\sigma_s$ = **± 1.0%** at *E* < **1.6 MeV**.

  MF=33, MT=1, $\mathbf{cov}_{tot}(i, i')$: 10^{-5} eV < E_1 < 0.1 MeV, 0.1 MeV < E_2 < 1.6 MeV, ...

Questions to be discussed (n scattering)





- In new **estimates of** ^{16}O: $\sigma_{s,\,th} \pm \Delta\sigma_{s,\,th}$, what will it be ? (*e.g.*, consistent with available **neutron optics data**, and σ_{tot} of **Dilg-1971** (E = 130.0 eV) & **Block-1975** (E = 23.5 keV) **OR** ...)

Energy bin structure for MF33: not too coarse **at low neutron energies**, e.g., a few bins at E < 1.0 MeV,

10^{-5} eV < E_1 < 0.01 MeV, 0.01 MeV < E_2 < 0.1 MeV, 0.1 MeV < E_3 < 1.5 MeV...

Then, **how accurately do we know σ_s (E) and σ_{tot} (E) at E_1 < 0.01 MeV: ~ ±0.1% or ~ ±1% ?**

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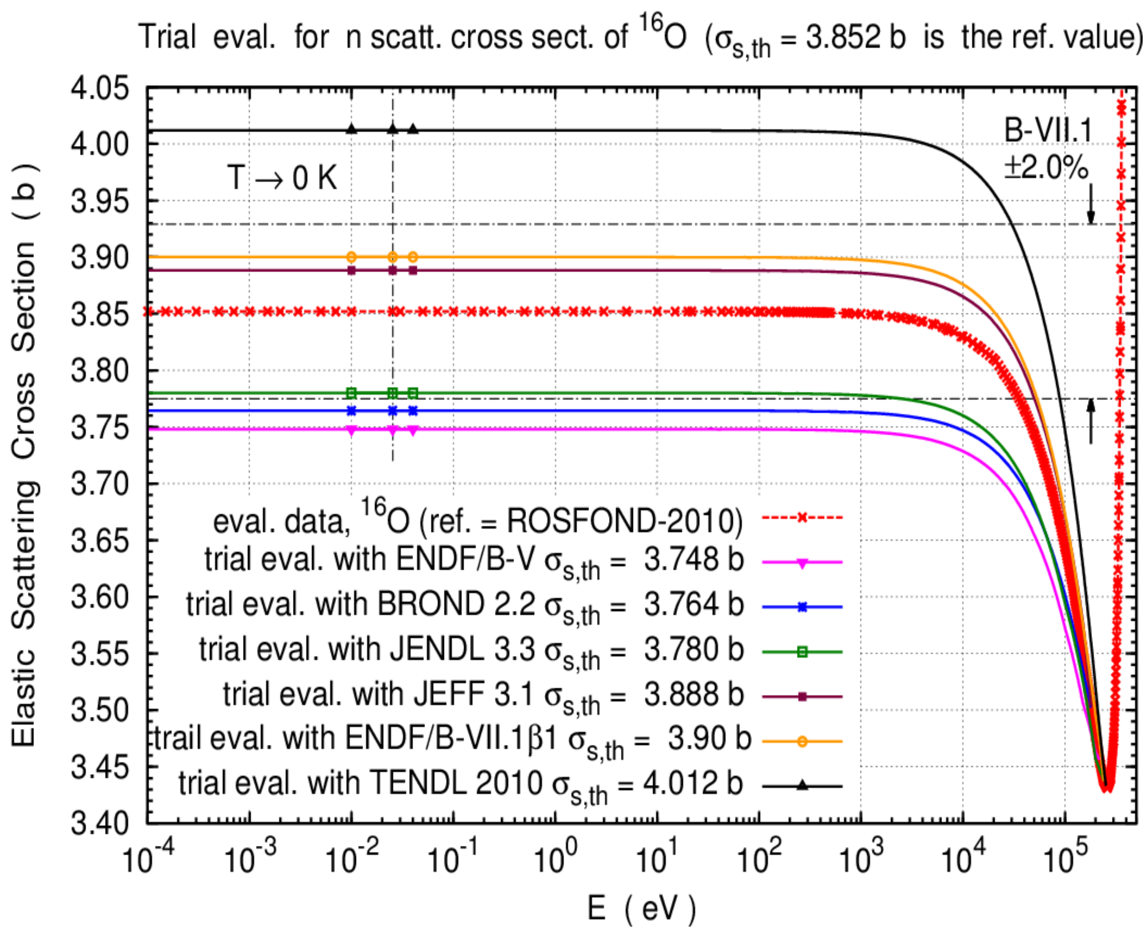
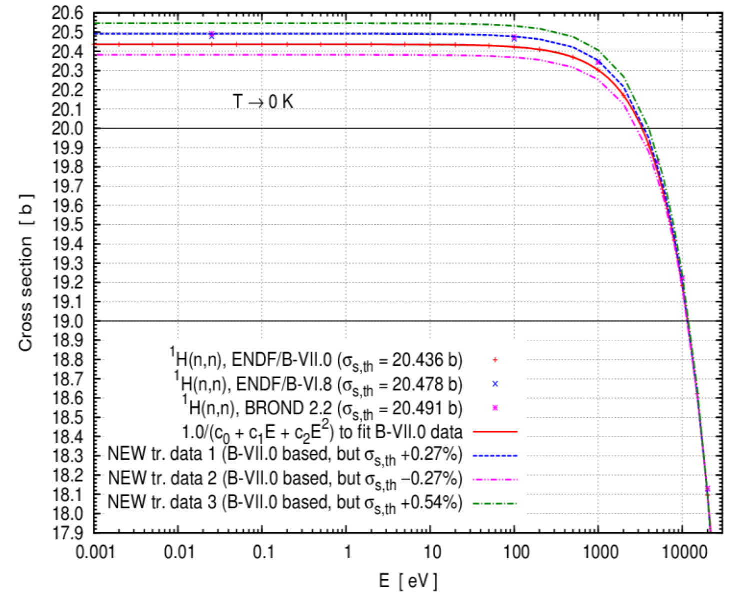
Uncertainty propagation (thermal n scatt.)





- We can answer the following question: for ^{16}O (as well as ^{1}H, ^{2}H) estimate the **uncertainty propagation** of $\sigma_{sc,\,th} \pm \Delta\sigma_{sc,\,th}$ (in MF3 and MF33 of an evaluation) to $k_{eff} \pm \Delta k_{eff}$ (of a critical benchmark) using **MCNP**, NJOY, and a linear regression:

$$k_{eff}(\sigma_{s,\,th}) \approx b + a \times \sigma_{s,\,th}(^{16}O) + a' \times \sigma_{s,\,th}(^{2}H) + \ldots$$

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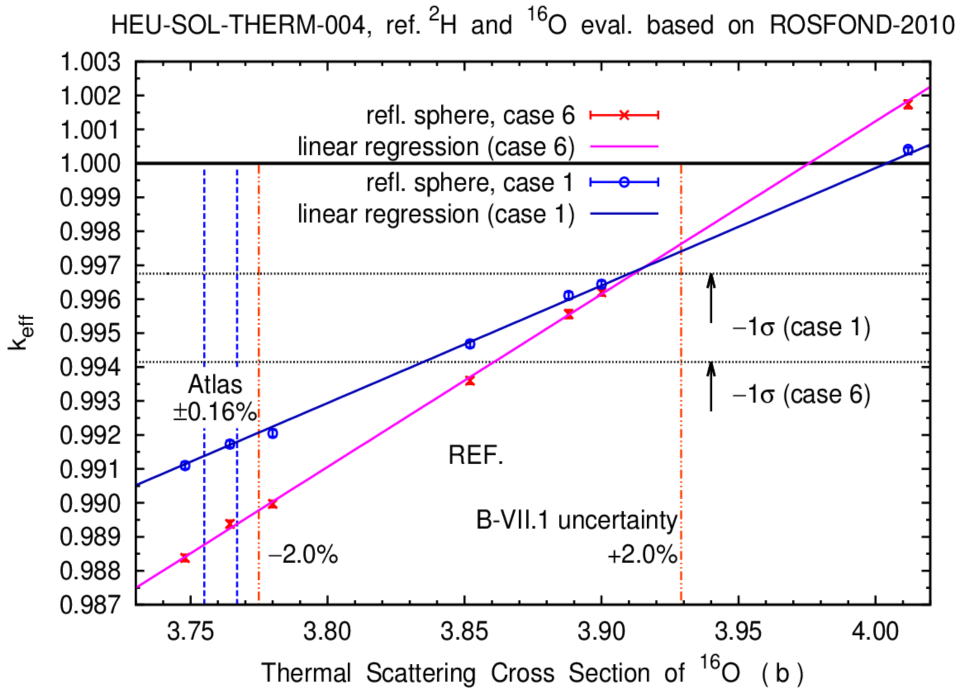
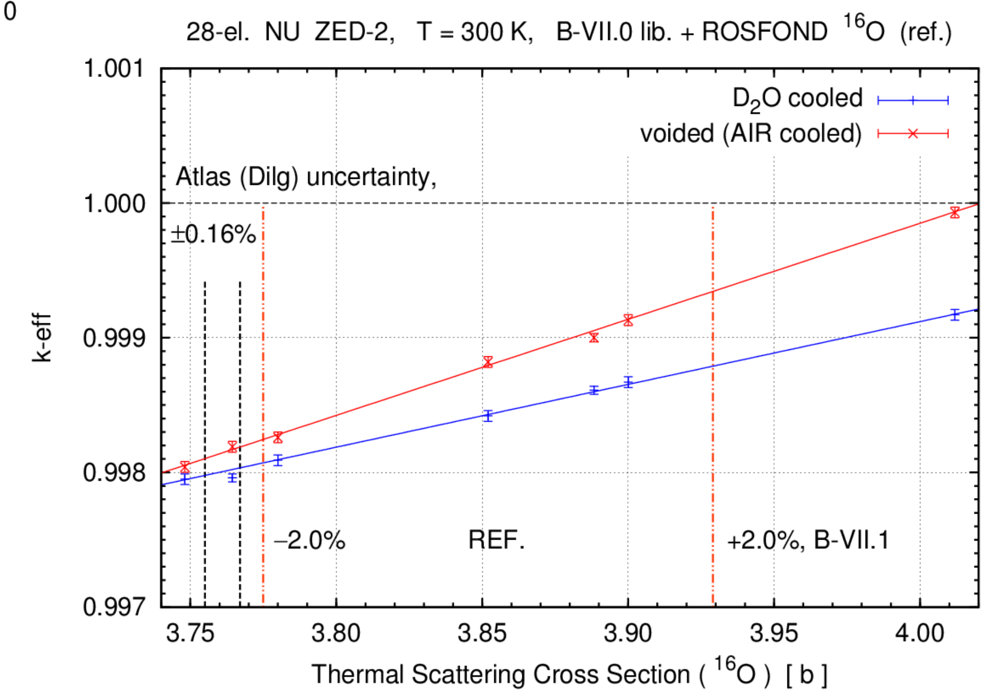
Using trial evaluations for $^{16}O(n,n)^{16}O$, we have





- $k_{eff}(\sigma_{s,\,th}) \approx b + a \times \sigma_{s,\,th}(^{16}O)$, **valid at** $\Delta\sigma_{sc,\,th}$ **~ ±1% ;**
- $S_{s,\,th} \propto \partial k_{eff} / \partial \sigma_{s,\,th} = a$ (sensitivity coefficient).
  We normalize it to be in ***mk per %*** **(1 mk = 100 pcm)**
- **Benchmark HST-04 (Case 6)**, $S_{s,\,th}(^{16}O) \approx 2.0$ mk per %, but $\Delta k_{eff} = \pm 5.85$ mk
- Typical **ZED-2** cores with NU fuel, $S_{s,\,th}(^{16}O) \approx 0.2$ mk per %, but $\Delta k_{eff} \approx \pm 3$ mk
  compare with $S_{s,\,th}(^{2}H) \approx 1.2$ mk per %, $S_{s,\,th}(^{1}H) \approx 0.4 - 0.5$ mk per %.

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References (^{16}O)

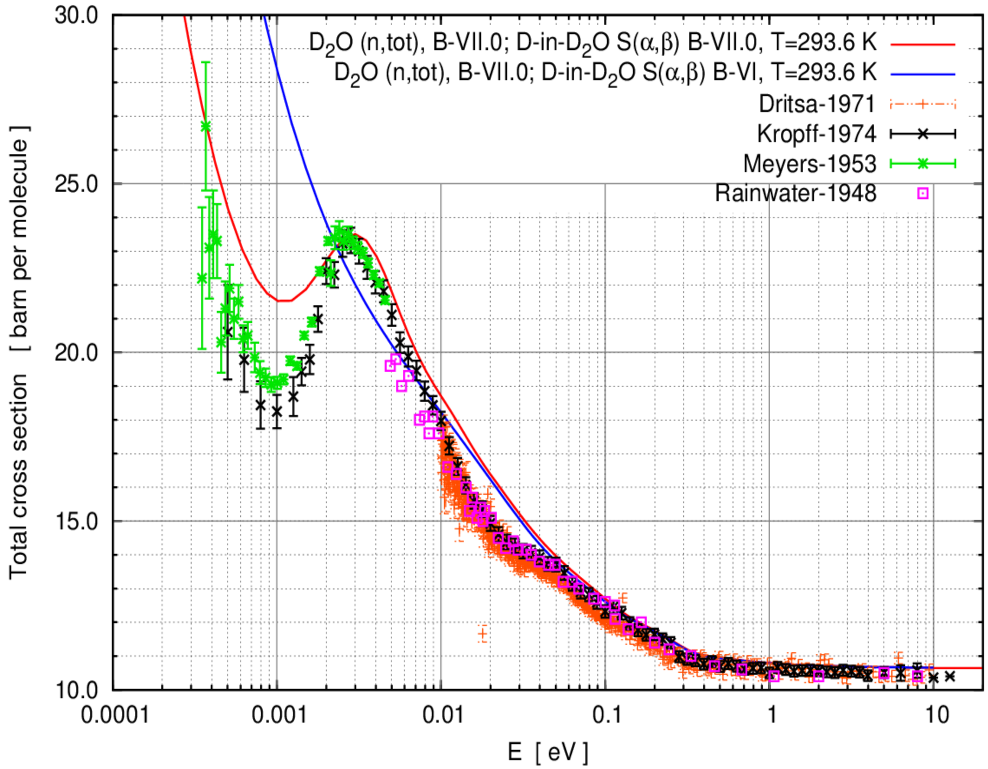
- A. Plompen, S. Kopecky, K. Kozier, D. Roubtsov, ***"The status of low energy data for deuterium, oxygen and hydrogen,"*** JEFF Document JEF/DOC-1488, Nuclear Energy Agency (NEA), Paris, 2013 (February),

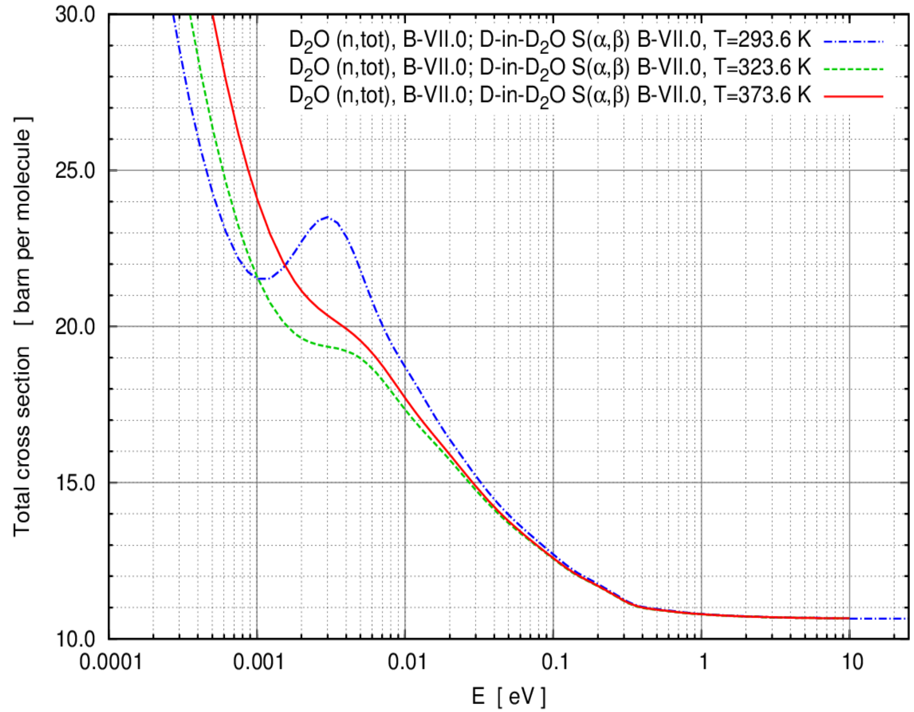
  http://www.oecd-nea.org/dbdata/jeff/jeffdoc.html

- K. Kozier, D. Roubtsov, A. Plompen, S. Kopecky, "***Reactivity Impact of ^{16}O Thermal Elastic-Scattering Nuclear Data for Some Numerical and Critical Benchmark Systems***," Nuclear Technology, Vol. 183, pp. 473-483, 2013 (September).

- K. Kozier, D. Roubtsov, J. Chow, A. Plompen, S. Kopecky, J. Svenne, and L. Canton, ***"Reactivity Impact of ^{2}H and ^{16}O Elastic Scattering Nuclear Data on Critical Systems with Heavy Water,"*** in Proceedings of ND-2013, Nuclear Data Sheets.

TSL: D_2O, and D_2O *vs.* H_2O





- **$\sigma_{tot}(E)$ in barn per molecule for D_2O ($^2H_2{}^{16}O$) in heavy water at *RT* and (T up & down)**
- **Near E_{th} = 0.0253 eV, we have at *RT***

(*Exp*. – *Calc*.) /*Calc*. ≈ –8.4% (using B-VII.0 **S(α,β)** for D-in-D_2O): **?**

TSL: H_2O and D_2O

With **J.I. Márquez Dámian**, **J.R. Granada**, *et al.*

(Centro Atómico Bariloche, Argentina),

the goal is

A NEW EVALUATION OF

THERMAL SCATTERING LAW

FOR LIGHT AND HEAVY WATER

IN ENDF-6 FORMAT,

BASED ON EXPERIMENTAL DATA

AND MOLECULAR DYNAMICS

WITH MODERN SIMULATIONS OF WATER DYNAMIC AND STRUCTURE

we will have ^{16}O (*new*), any (reasonale) *T* available,

etc.

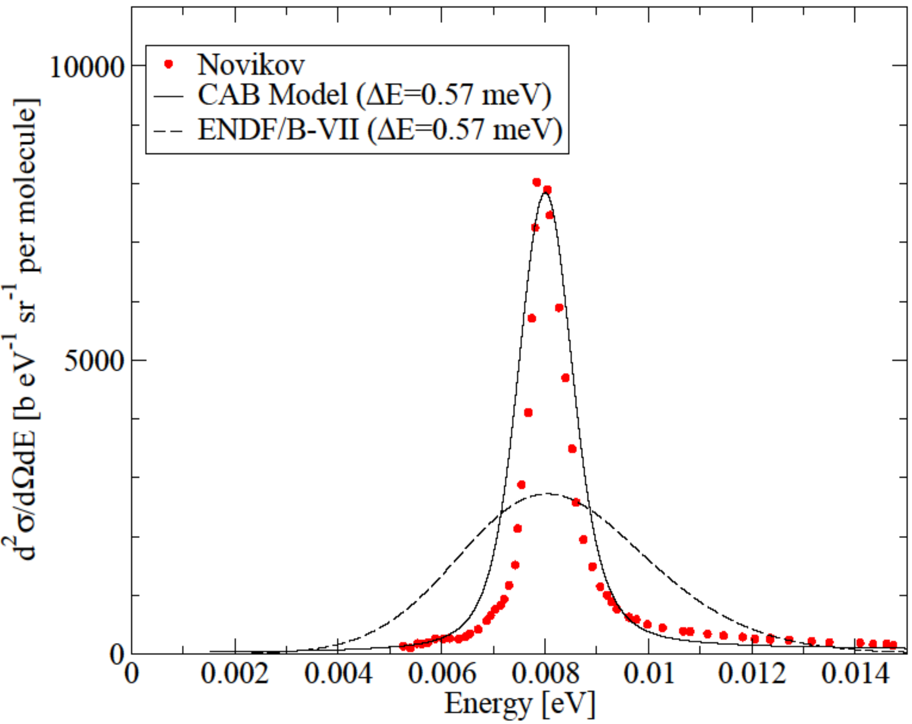
S(α,β) for water: collaboration with Bariloche

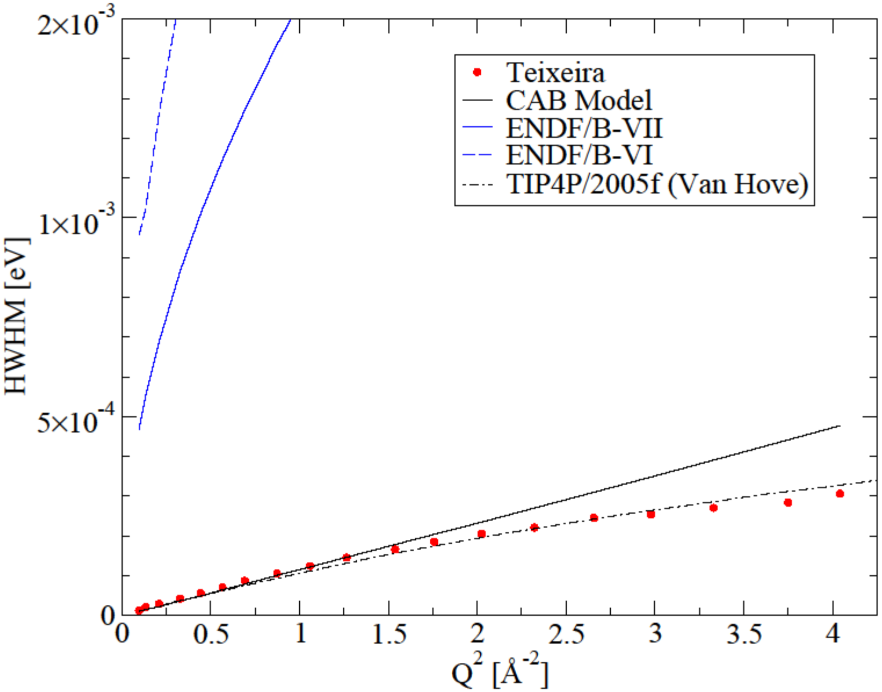
- The Neutron Physics Department at *Centro Atómico Bariloche* in Argentina is working on an update of the thermal scattering libraries for light and heavy water. There are two researchers involved (J.R. Granada and J. Dawidowski) , one former post-doc (A.D. Viñales) , and one PhD student (J.I. Márquez Damián).
- Their approach is based on combining **molecular dynamics** simulations and experimental data, and the resulting models are implemented in **LEAPR** / NJOY.
- The **key points** for these models are:

1. use of molecular diffusion for translational motion (instead of free gas approx.),
2. continuous spectra computed from molecular dynamics simulation at a given thermodynamic state (instead of derived from neutron scattering experiments),
3. Models for D and O in D_2O, with a more precise description of **the structure** (instead of the incoherent approximation used in ENDF/B-VI or the Lennard-Jones model for D-D structure used in ENDF/B-VII).

- The resulting cross sections are an improvement over existing evaluations: **they are compared with measurements** of double differential scattering cross sections, quasi-elastic neutron scattering measurements, angular distributions, average cosine of the scattering angle and total cross sections.

Results: light water TSL (1)





Double differential scattering cross section for light water, E_0 = 8 meV, θ = 37°.

Half-width of the quasi-elastic peak for E_0 = 3.15 meV neutrons in H_2O.

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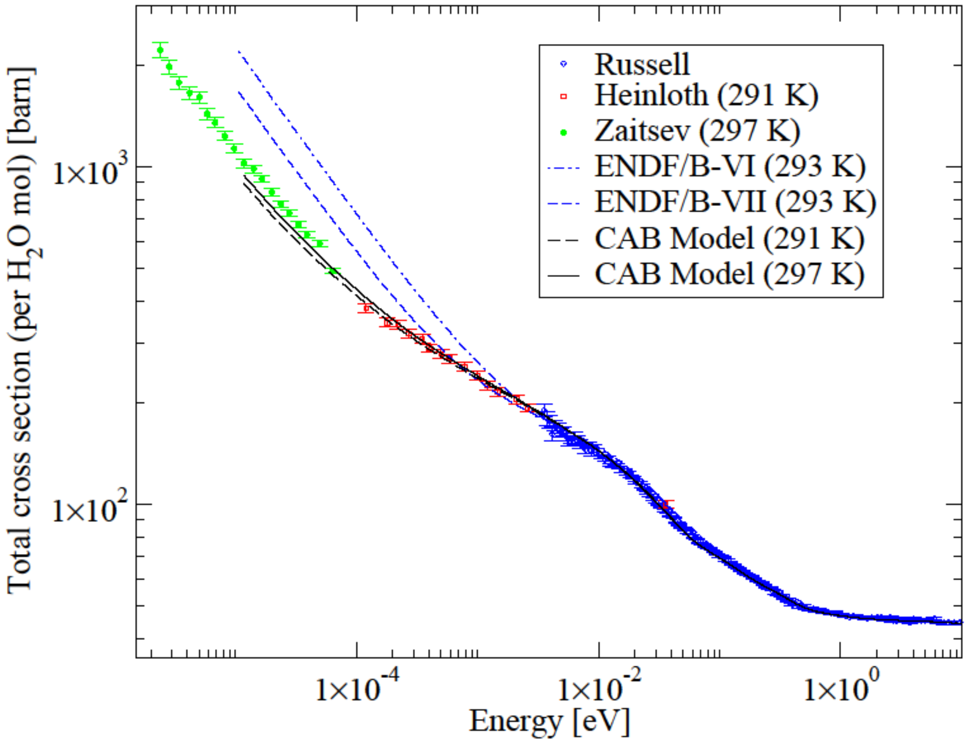
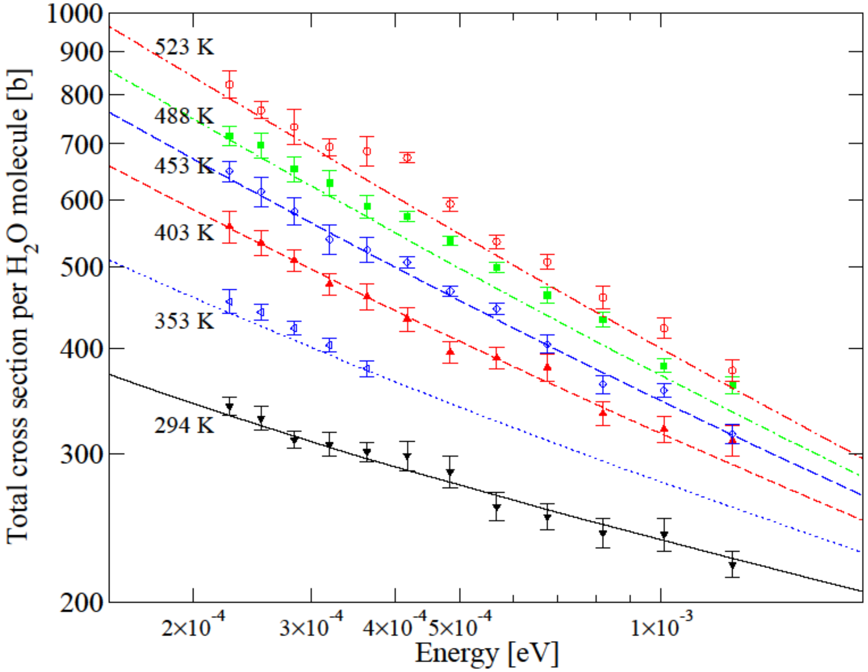
Results: light water TSL (2)





Total cross section
per H_2O molecule (RT).

Temperature dependence
of the total neutron cross section
for H_2O.

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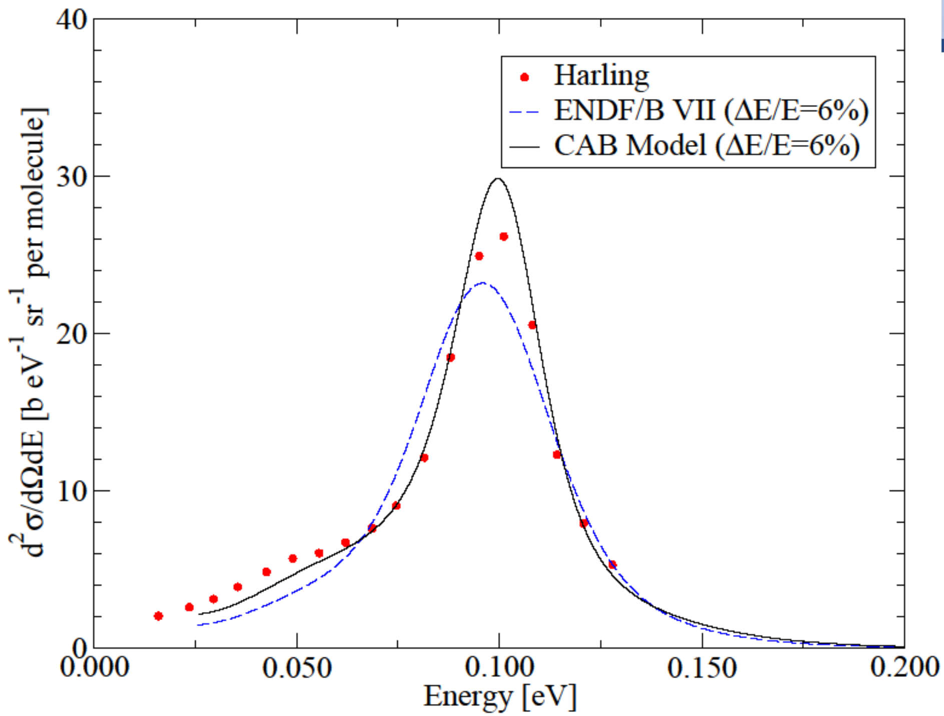
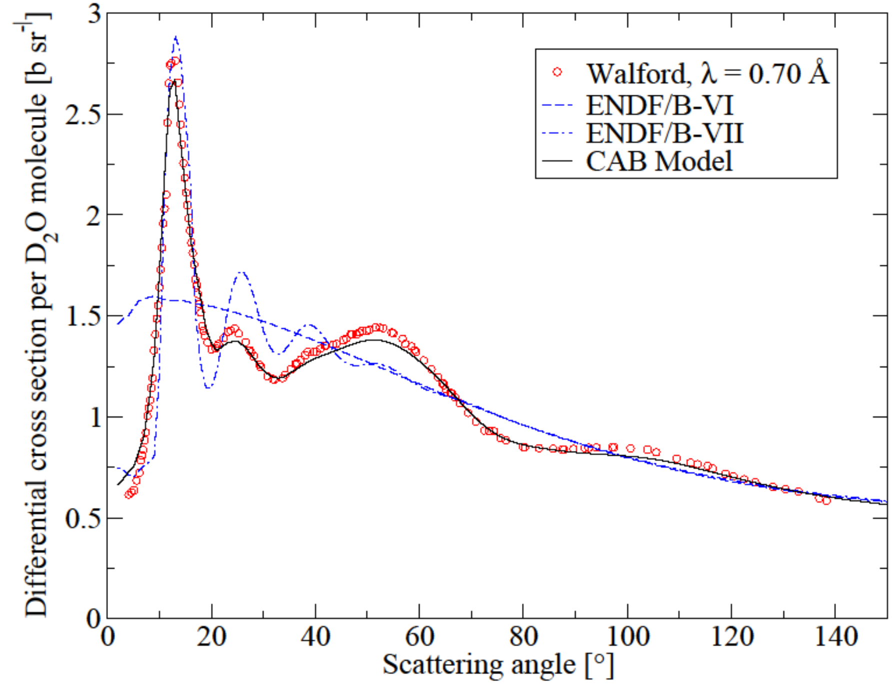
Results: heavy water TSL (1)



Double differential scattering cross section for heavy water, E_0 = 101 meV, θ = 60°.



Angular distribution for E_0 = 170 meV neutrons in D_2O.

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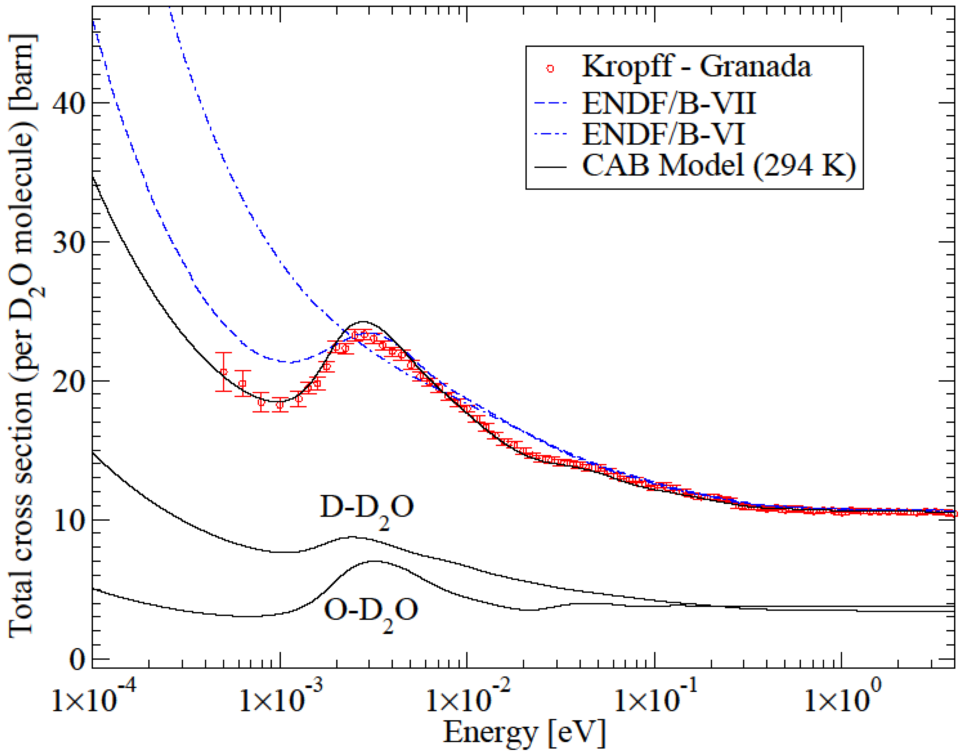
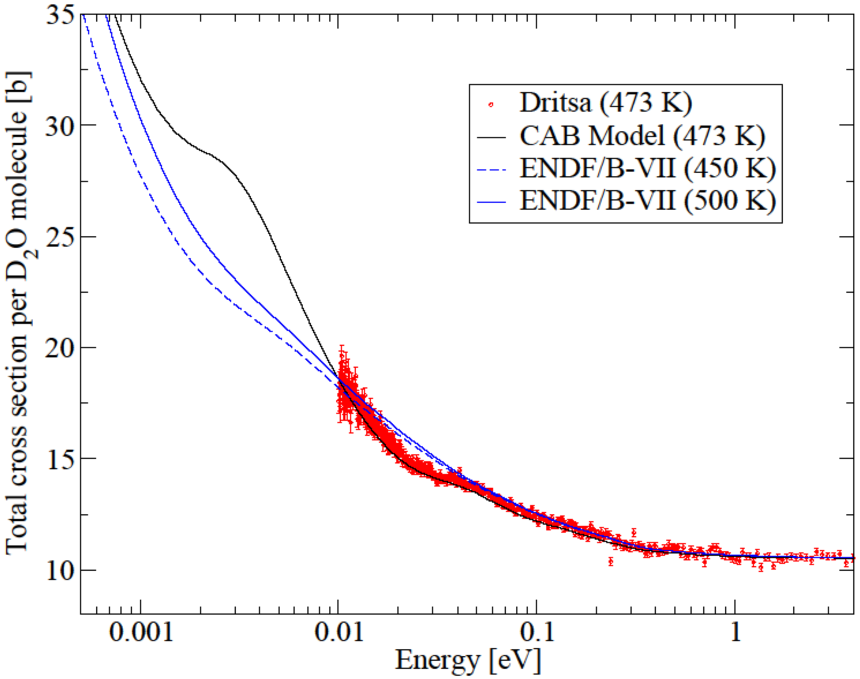
Results: heavy water (2)





Total cross section
per D_2O molecule (at RT).

Total cross section
per D_2O molecule at T = 473 K.

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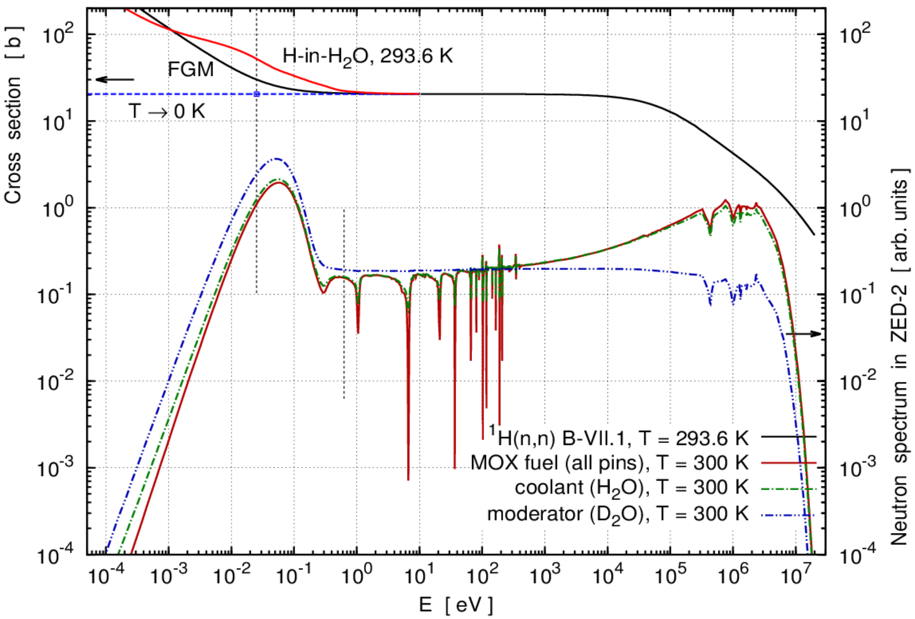
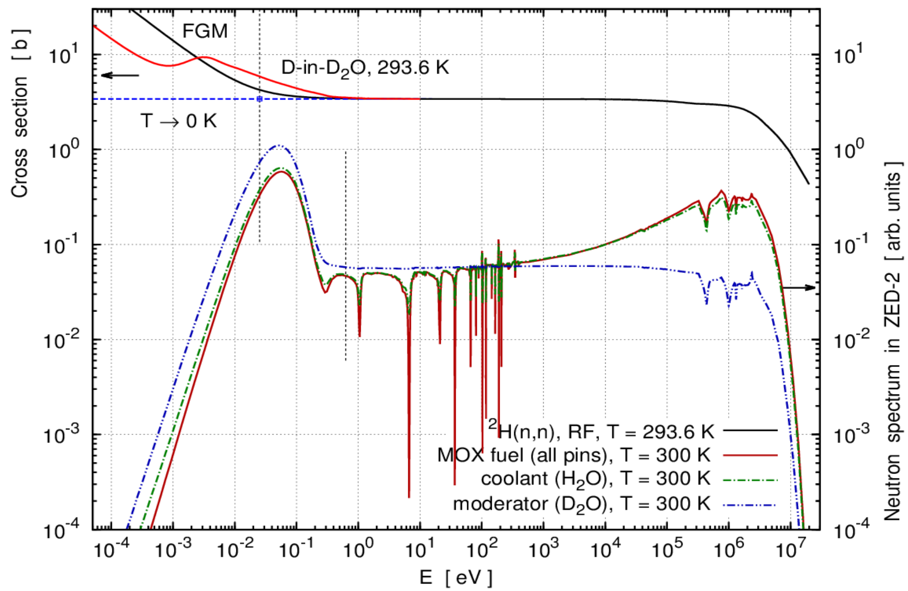
Testing New TSL (thermal ace + MCNP5)





We create new thermal ace files (at room T) with NJOY99 and use MCNP5 for critical benchmarks with H_2O and D_2O ($\boldsymbol{k}_{\mathbf{eff}} \pm \Delta \boldsymbol{k}_{\mathbf{eff}}$)

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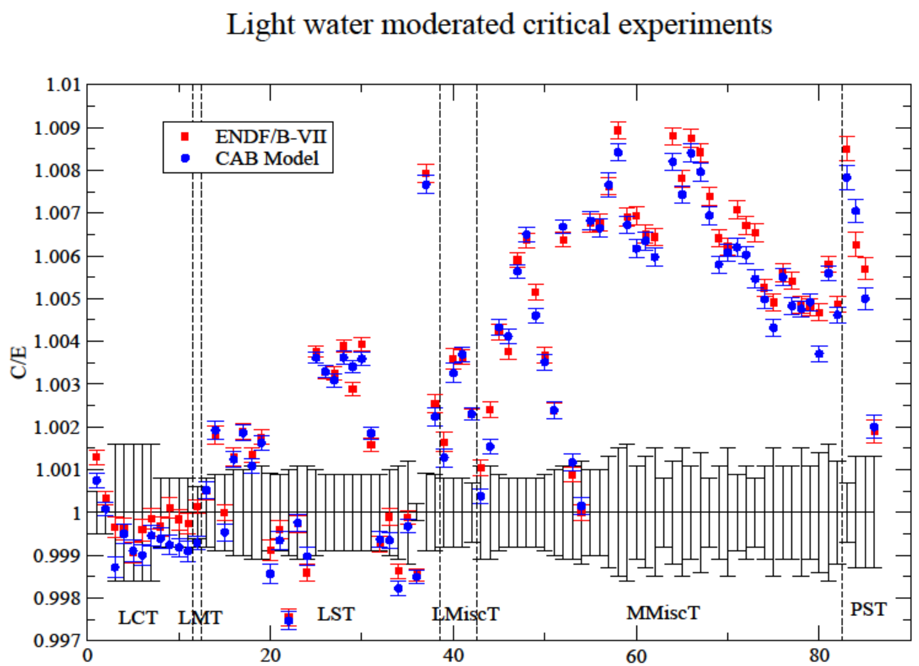
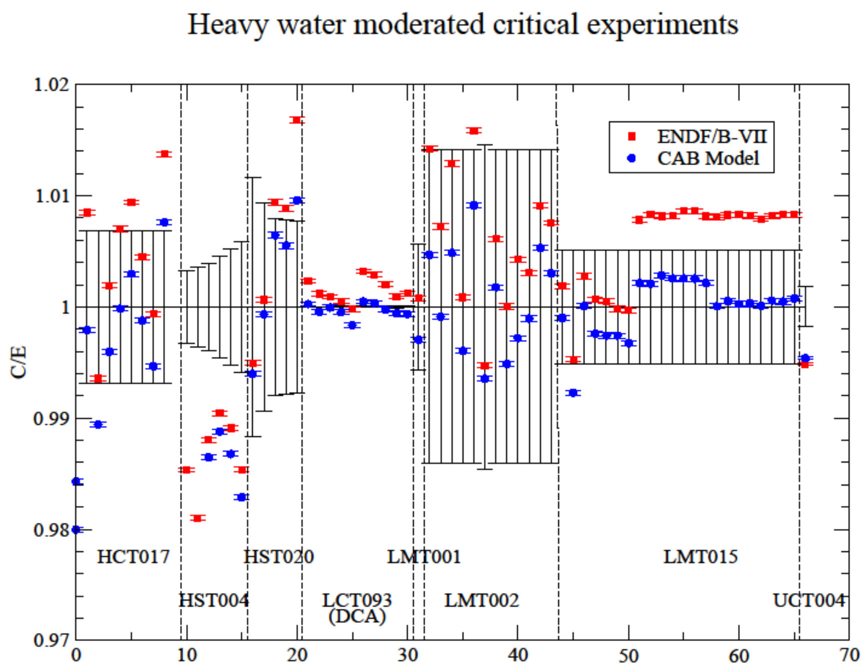
Results: ICSBEP criticality benchmarks (C/E ratio)





ENDF/B-VII + ENDF/B-VII TSL *vs.* ENDF/B-VII + CAB Model (H_2O)

- Impact is less than 100 pcm.
- No clear improvement over ENDF/B-VII TSL.
- Existing discrepancies caused by nuclear data not addressed.

ENDF/B-VII + ENDF/B-VII TSL vs. ENDF/B-VII + CAB Model (D_2O)

- Differences up to 1000 pcm.
- 66% of benchmarks cases have a C/E ratio closer to 1.0 using the new evaluation.
- 82% when the new evaluation and ^{2}H (fast ace) from ROSFOND-2010 is used.

Summary for new evaluations of TSL (water)

- The ***CAB Models for water*** are a set of new models for thermal neutron scattering in light and heavy water, generated from molecular dynamics calculations and experimental data.
- The models result on a improvement over existing models for both light and heavy water for the calculation of differential and integral scattering quantities.
- The impact of the improvements in light water is not significant for criticality applications (main differences are in low energy, small angle interactions), but updating the library might be important for other users.
- In the case of heavy water, the inclusion of O-in-D_2O and a better representation of the structure improves the calculation of critical systems significantly.
- **REFERENCES**

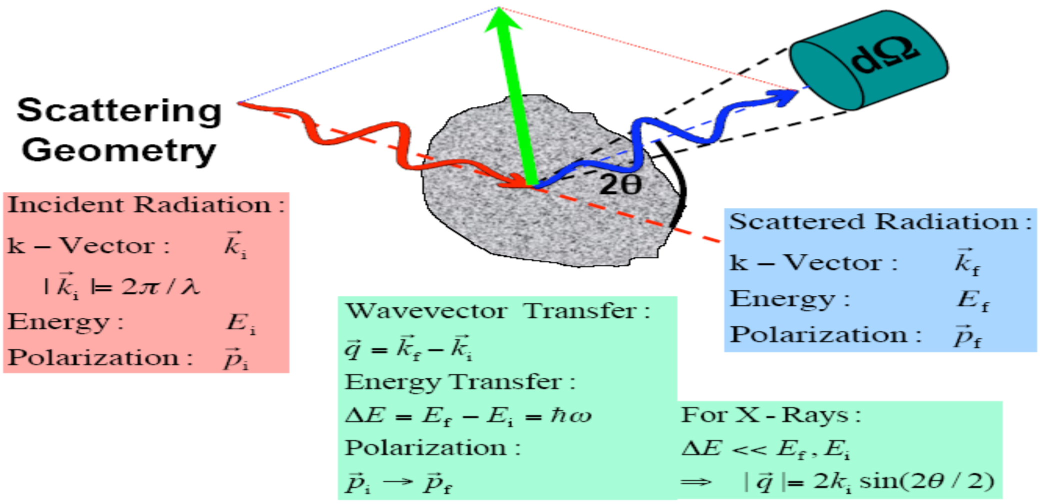
J.I. Márquez Damián, J.R. Granada, D.C. Malaspina, Ann. of Nucl. En. (accepted), **65**, (2014).

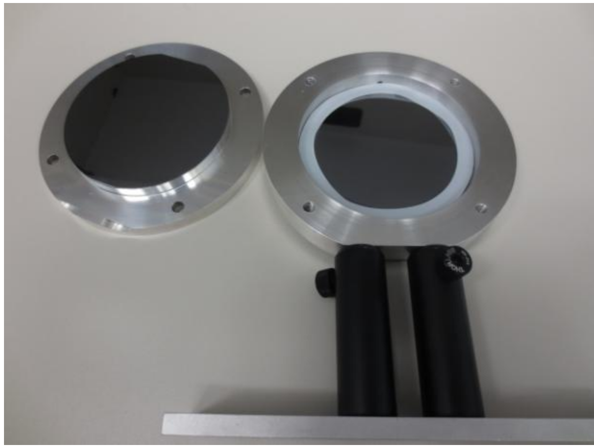
J.I. Márquez Damián, D.C. Malaspina, J.R. Granada, J. Chem. Phys., **139**, p. 024504 (2013).

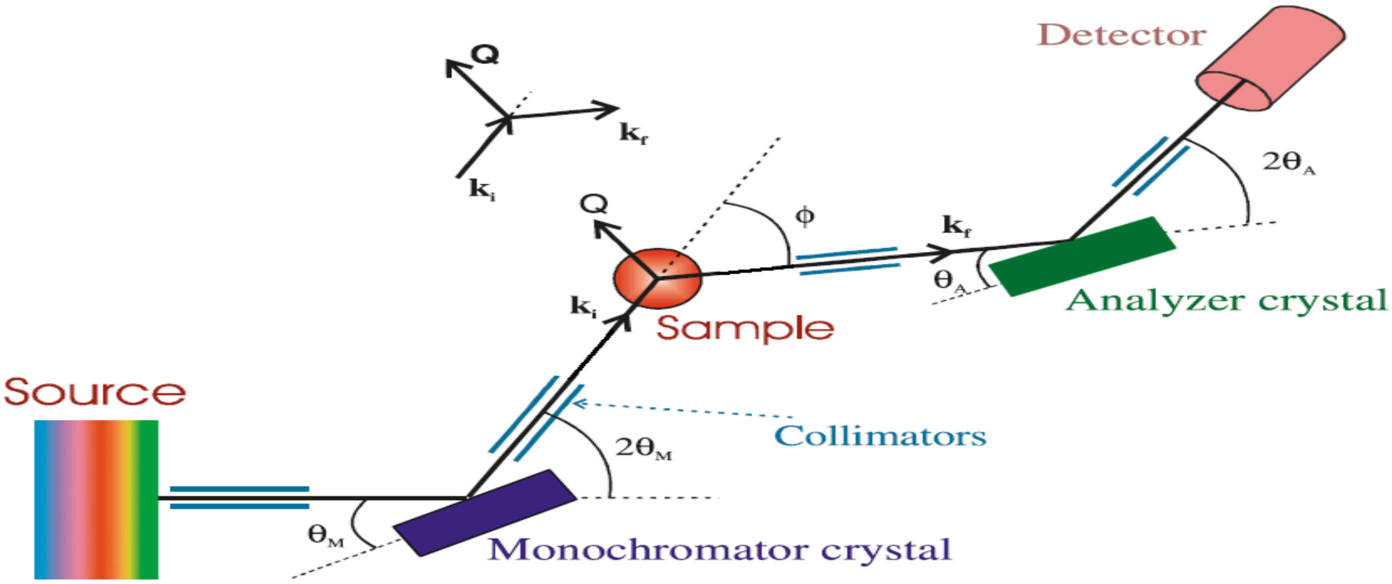
A.D. Viñales, J. Dawidowski, J.I. Márquez Damián, Ann. of Nucl. En., **38**, p. 1687 (2011).

J.I. Márquez Damián, J.R. Granada, D.C. Malaspina, in Proceedings of **ND-2013**

Neutron Scattering on Water using triple axis spectrometer at NRU reactor at CRL, AECL







Sample sizes: 0.1 mm to 3 mm, at RT

Triple axis spectrometer

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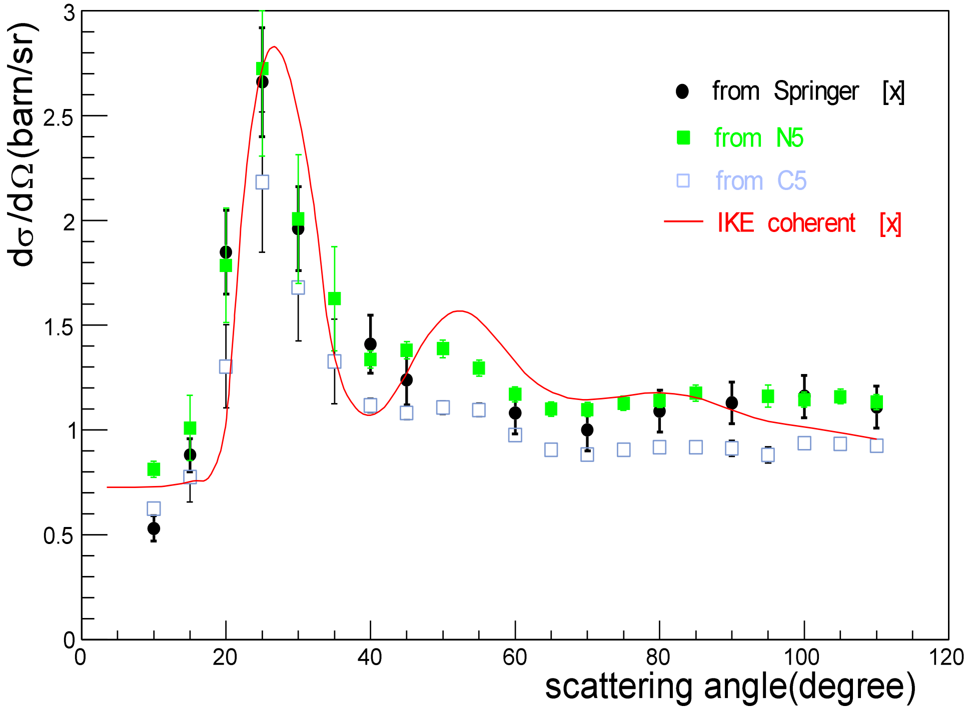
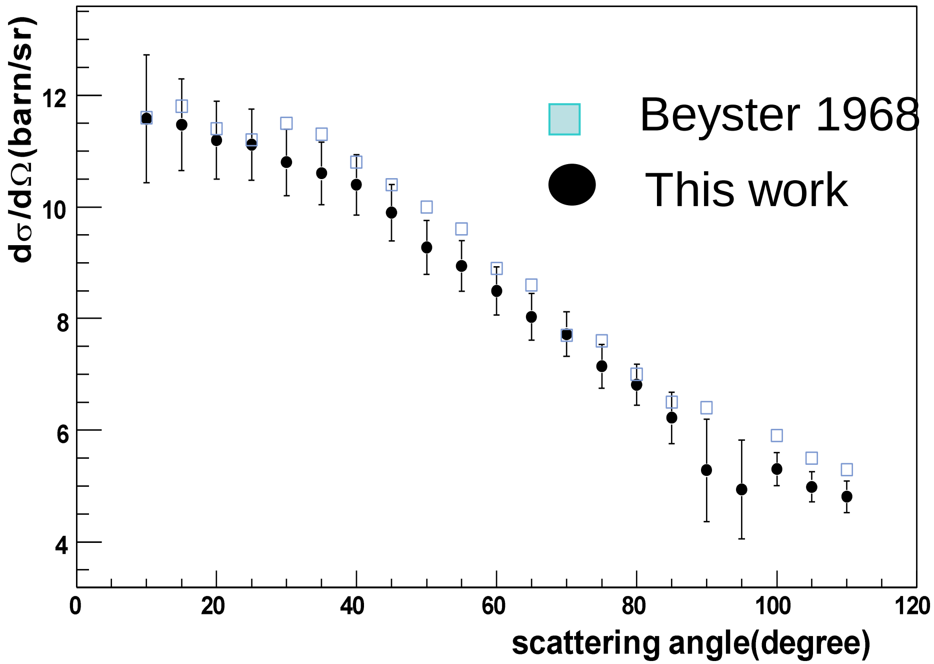
G. Bentoumi, G. Li, B. Sur, and Z. Tun, (AECL and Canadian Neutron Beam Centre)





Differential scattering cross section of D_2O.
Beam energy is 44 meV
(except for that the result from C5: it is at E_0 = 41.44 meV).

Absolute differential cross section (in barn/sr)
for Light water,
at E_0 = 41 meV,
Beyster's data: ±5%.