IRSN
INSTITUT
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Testing thermal scattering law for light water at 600 K using VESTA 2.2.0 depletion calculations

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OUTLINE

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- VESTA simulations
- Results
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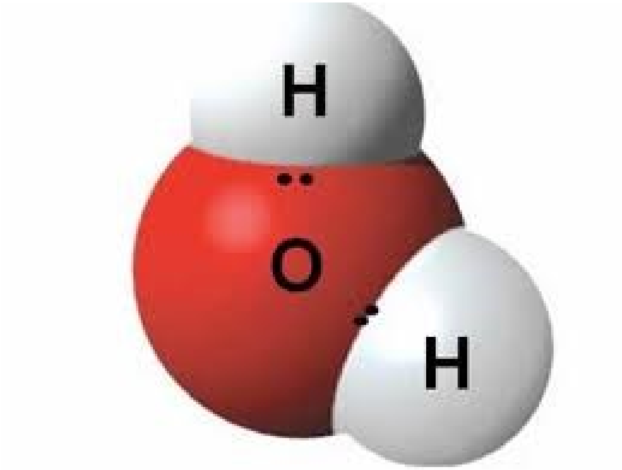
  - Validation results
- Conclusions

Motivations and new $H(H_2O)$ TSL



Motivations

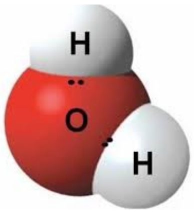
- To improve the design and safety of critical systems and thermal reactors, it is necessary to:
  - perform high-precision neutron transport calculations
  - use state-of-the-art nuclear data

- For reactors applications: light water is the most common moderator

- Thermal scattering cross-section data often termed as Thermal scattering law (TSL) for light water **play a major role !**

- Recent release of the US ENDF/B-VIII.0 library included H_2O reviewed version of TSL data based on Molecular Dynamics (MD) simulations whereas JEFF ones are based on old experiments

Motivations



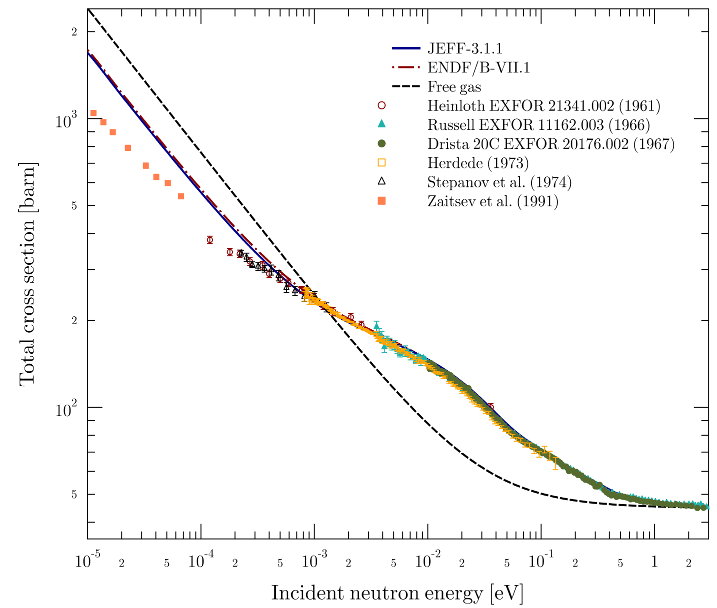
- For room temperature: they perform well, but temperatures other than room temperature are needed for reactor physics!

- At IRSN, an effort has been devoted to the investigation and evaluation of $H(H_2O)$ TSL for temperatures higher than room temperature for reactor safety and criticality safety applications (BUC). For so, thermal neutron scattering cross section for hydrogen bound in H_2O was re-evaluated at 600 K based on MD simulations

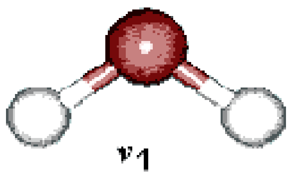
- In this work, we present the impact of the new $H(H_2O)$ TSL data for PWR using VESTA 2.2.0 depletion calculations, by comparing the results obtained using both ENDF/B-VIII.0 $H(H_2O)$ TSL and the new IRSN TSL data

New IRSN $H(H_2O)$ TSL

- In ENDF/B-VIII.0: a new TSL library based on the **CAB model** has been adopted. This new library involves **MD simulations** of light water using the **non-polarizable TIP4P/2005f flexible light water potential.** This methodology is not completely approximation-free, due to the intrinsic limitation of the classical MD simulations.



- At IRSN: $H(H_2O)$ TSL was recently re-evaluated at reactor operating temperature, i.e., at 600 K based on MD simulations using the **PolarisMD code** and **TCPE polarizable rigid water potential.**



This potential takes into account the **effect of polarization of the water molecules**, believed to be an important parameter while defining a water potential at high temperature.

V. Jaiswal thesis, 2018

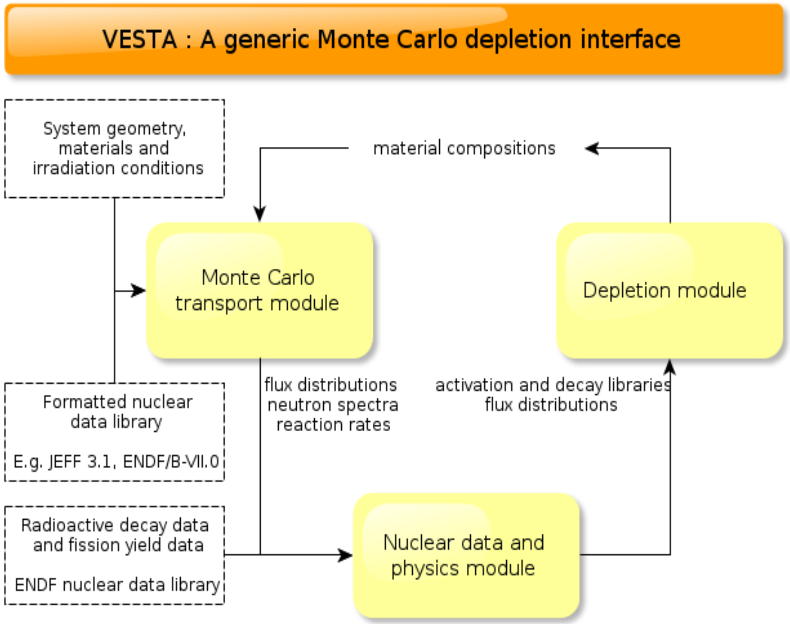
VESTA simulations

VESTA : Monte-Carlo depletion code

- Goal: accurately simulate material composition under irradiation

- Applications: criticality safety, reactor safety studies (PWR, BWR, research reactors, fusion systems, ..), cycle (transport, scenario ..), waste characterization, material activation ..

- VESTA philosophy: a generic interface customisable to a user's needs:
  - Monte Carlo code: **MCNP**(X/**6**), MORET 5
  - Depletion module: ORIGEN 2.2, **PHOENIX**, FISPACT,..
  - Nuclear data libraries: JEFF, JENDL, ENDF/B at different temperatures



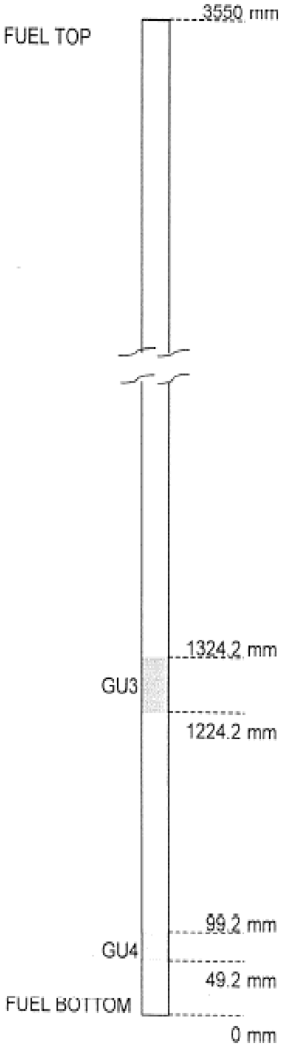
- VESTA 2.1.5 has been validated for 65 isotopes on 96 radiochemical assay and decay heat measurements using JEFF-3.1 and ENDF/B-VII.0

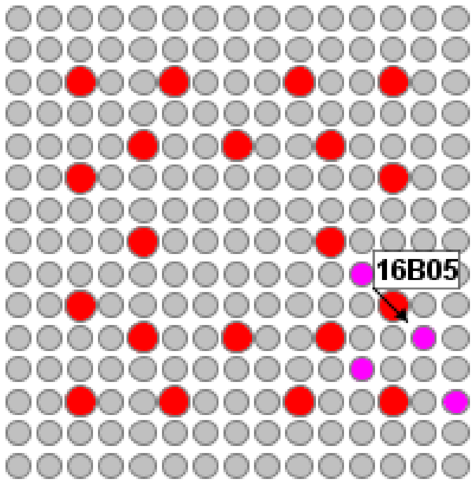
- VESTA 2.2.0 validation is ongoing using JEFF-3.2 and ENDF/B-VII.1

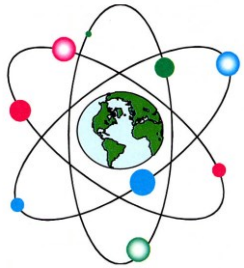
The ARIANE GU3 sample

- The ARIANE (Actinide Research In A Nuclear Element) program examined irradiated MOX and LEU fuel samples in both commercial PWR and BWR power reactors.

- The ARIANE.GU3 sample:
  - PWR UO_2 sample
  - $^{235}U/U$: 4.1%
  - Estimated BU = 52.5 MWd $kgHM^{-1}$.
  - 3 cycles in the Gösgen PWR in Switzerland between 1994 and 1997
  - Extracted from the original assembly after 2 cycles and inserted into another fresh assembly for the 3rd cycle.
  - Analyzed by 2 labs: ITU and SCK-CEN



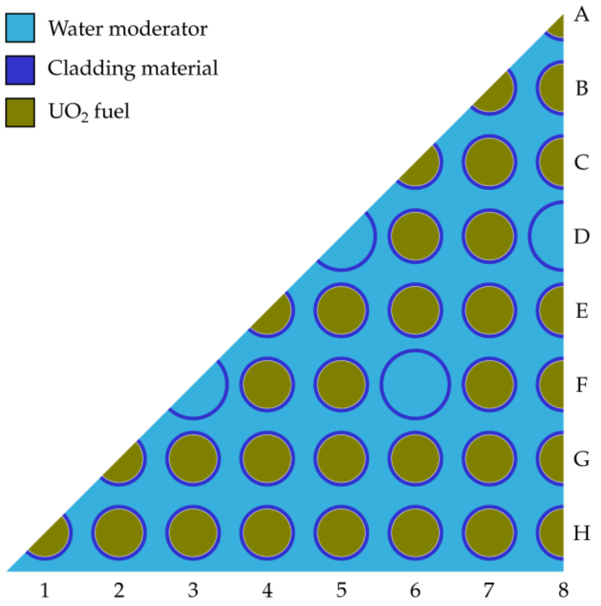




Model characteristics

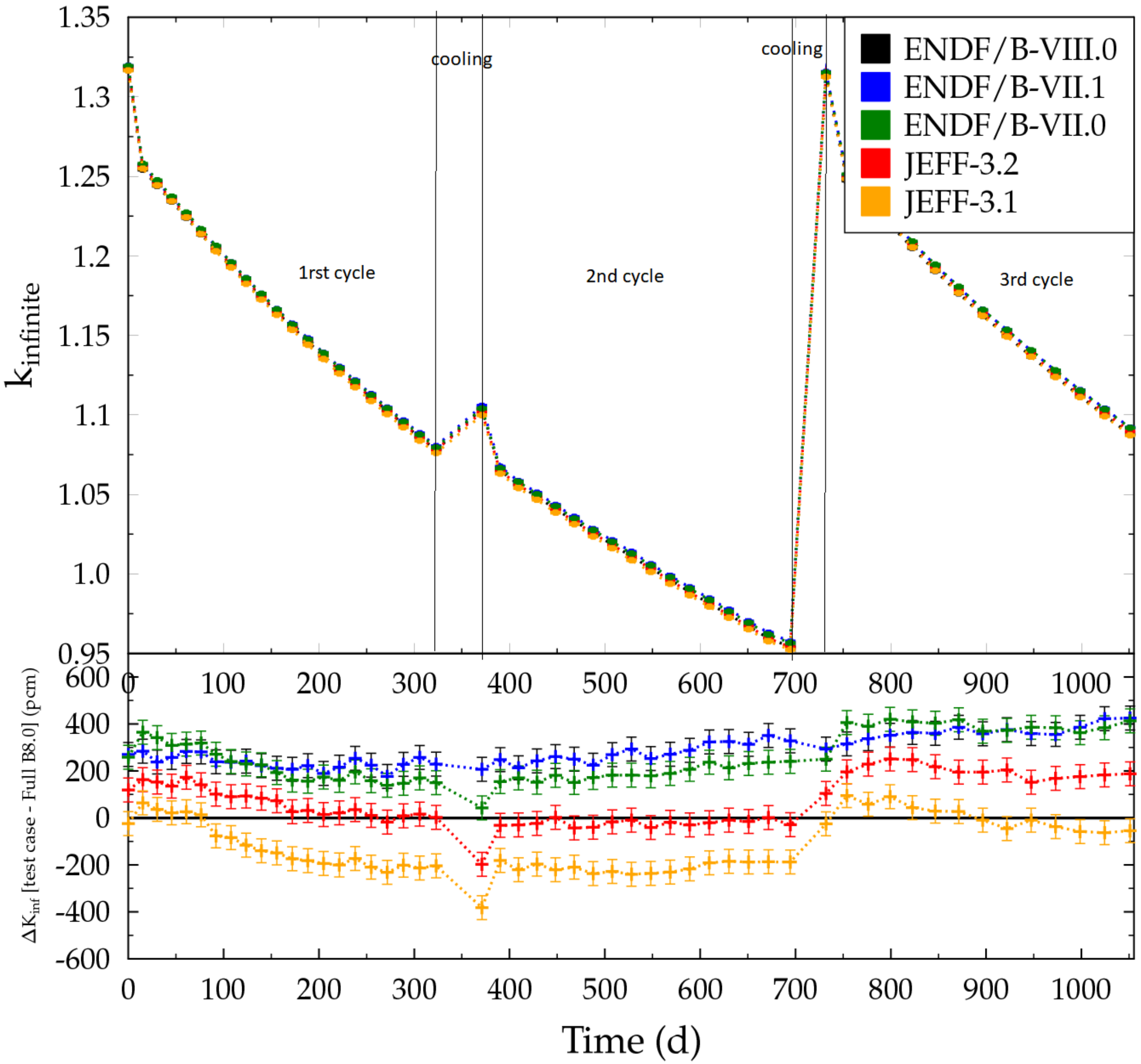
General model characteristics:

- 2D model – infinite medium
- 1m active height
- Average boron concentration
- Individual pins are being depleted (32 zones)
    - No radial zones for normal UO_2 or MOX fuel pins
- Model temperature:
    - Tmoderator (water) = 600 K
    - Tclad = 600 K
    - Tfuel = 900 K
- Nuclear data
    - Different JEFF & ENDF/B libraries
    - w/wo new IRSN $H(H_2O)$
- 53 BU steps of 1 $GWd.tHM^{-1}$



Results

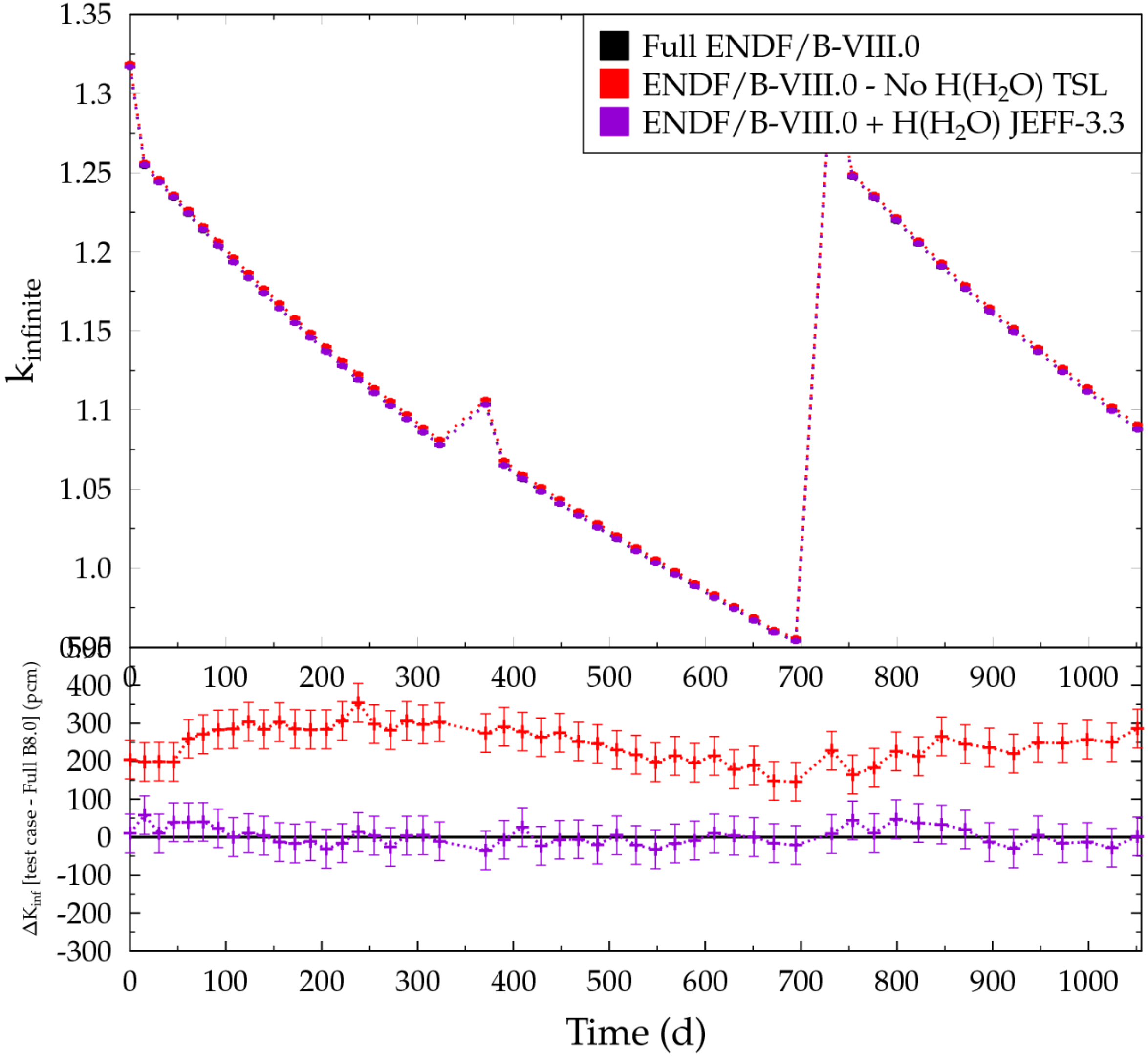
K_{inf} results



- Quite large discrepancies on k_{∞} results.
- ENDF/B-VII.1 and ENDF/B-VII.0 are overestimating B8.0 results by 200-400 pcm.
- JEFF-3.1 is in better agreement with B8.0 for the BOL, and is underestimating B8.0 by around 200 pcm for longer irradiation times.
- JEFF-3.2 overestimates B8.0 at BOL by 100-200 pcm, and is in very good agreement with B8.0 for most irradiation times.

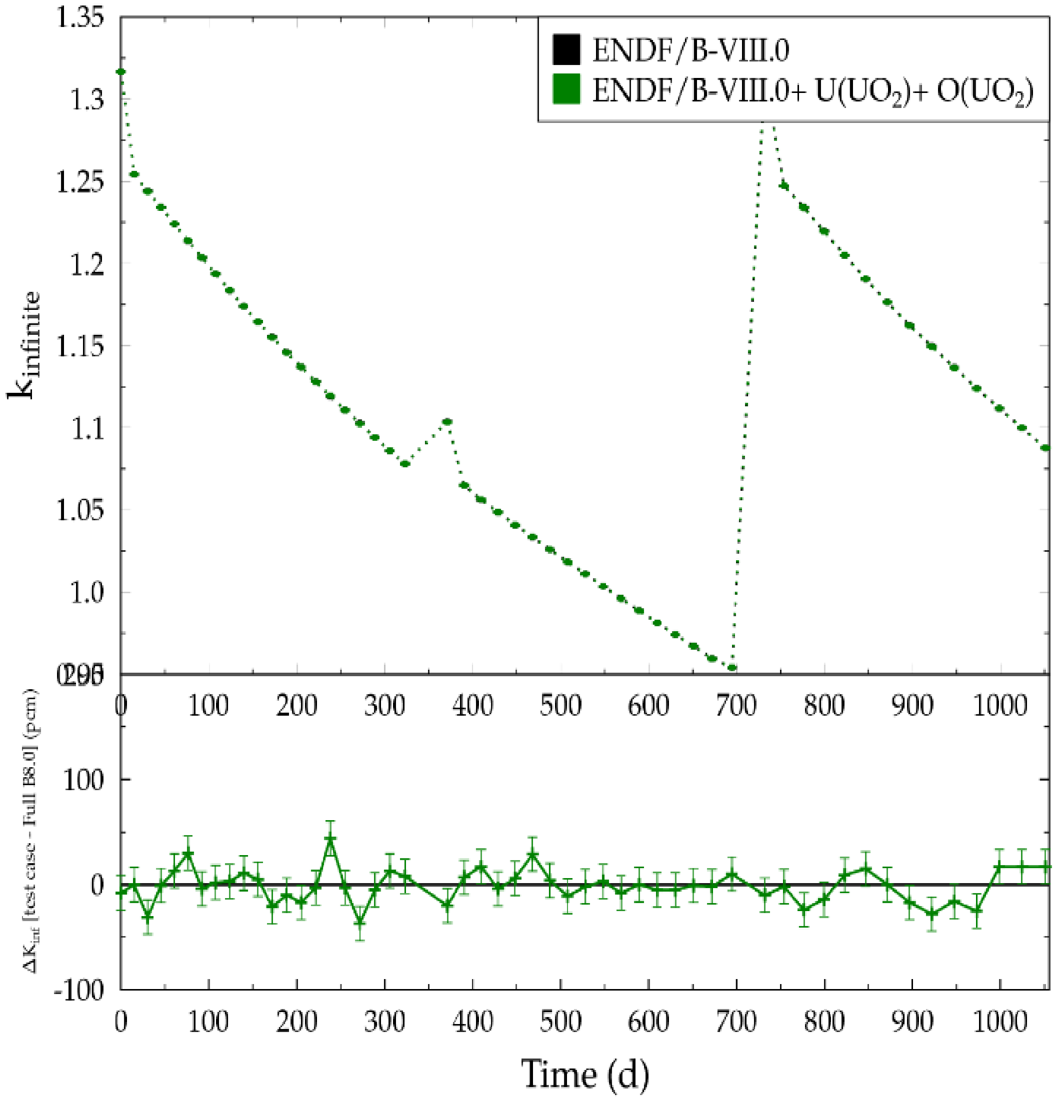
- The maximum discrepancy, goes up to 800 pcm (ENDF/B-VII.0 and JEFF-3.1)

K_{inf} results



- Impact of using any TSL data: up to 300 pcm during irradiation
- The recent $H(H_2O)$ TSL at 600 K in B8.0, compared to TSL from JEFF-3.3 do not show any significant impact during irradiation

K_{inf} results – w/wo U-O(UO_2) TSL

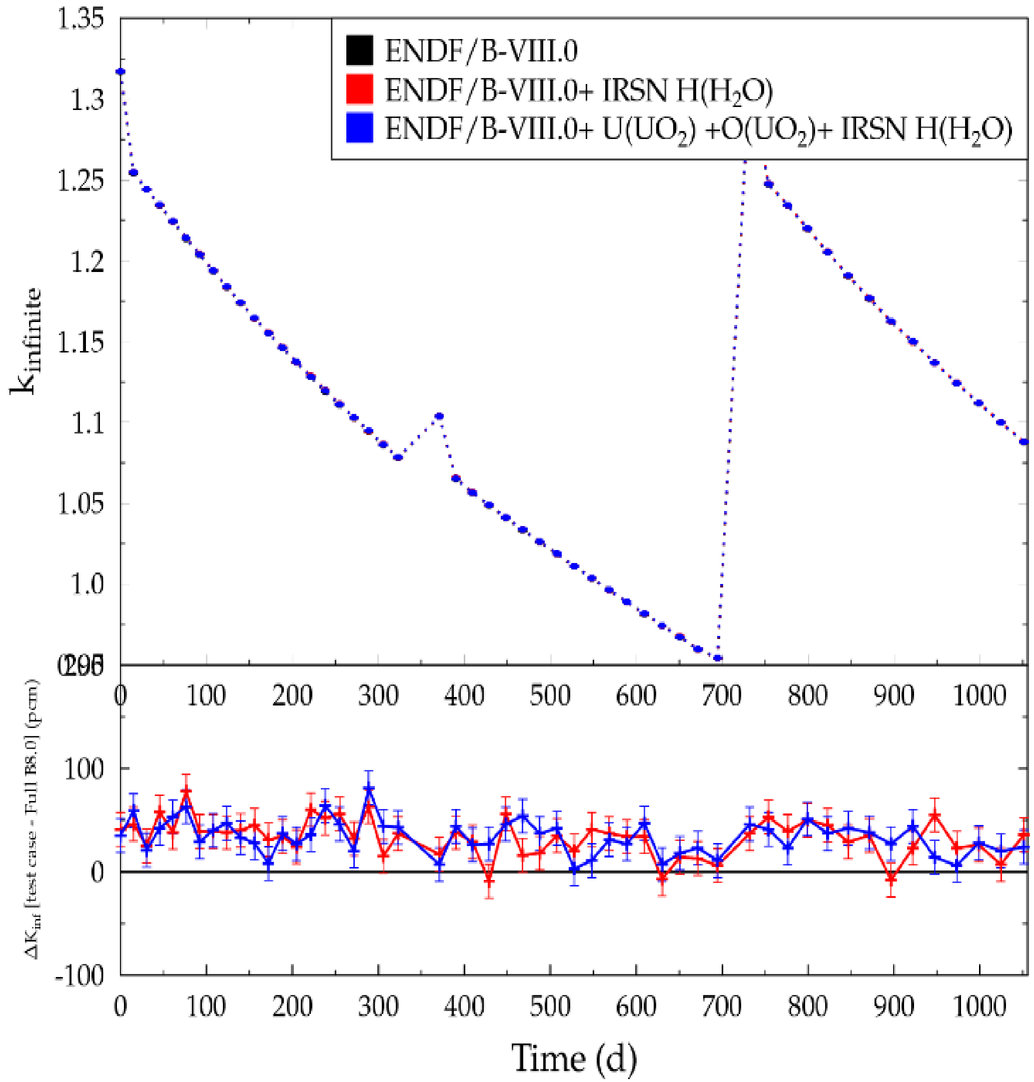


TSL for UO_2 has been evaluated in ENDF/B-VIII.0 using modern Ab Initio Lattice Dynamics (AILD) techniques. Neutron thermalization in UO_2 fuel may be impacted by the crystal binding effects of oxygen, the calculation of which may be facilitated with the use of TSL.

The partial U(UO_2) and O(UO_2) densities of states were used in the LEAPR module of NJOY2016.35 to generate TSL for U(UO_2) and O(UO_2) at 900 K for use in our model.

The new TSL for UO_2 at 900 K in the ENDF/B-VIII.0 data library do not show any significant impact during irradiation

K_{inf} results – $H(H_2O)$ IRSN TSL



The k_∞ results obtained when using:

- the new IRSN $H(H_2O)$ TSL
- the new IRSN $H(H_2O)$ TSL combined to the $U(UO_2)$ and $O(UO_2)$ TSL

show a slight over-estimation of roughly 30-50 pcm during irradiation, compared to full ENDF/B-VIII.0.

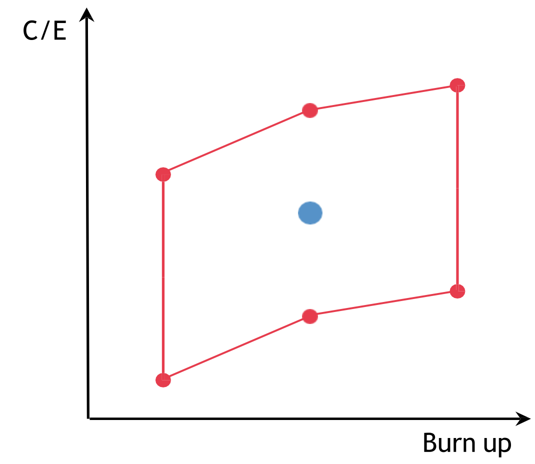
This indicates a slight impact of the new IRSN $H(H_2O)$ TSL at 600 K obtained with MD simulations and TCPE polarizable rigid water model potential.

MEMBER OF ETSON

Validation results

ENDF/B-VII.1 library

- For applications in the neutronics and criticality safety field, experimental validation is paramount.
- Provide a set of **C/E values** for the measured nuclides
- Define an **uncertainty range** for every nuclide, which includes:
  - the measurement uncertainty
  - the uncertainty introduced due to the irradiation history calibration (the combined value of ^{145}Nd, ^{146}Nd, and ^{148}Nd burn up tracers is applied)



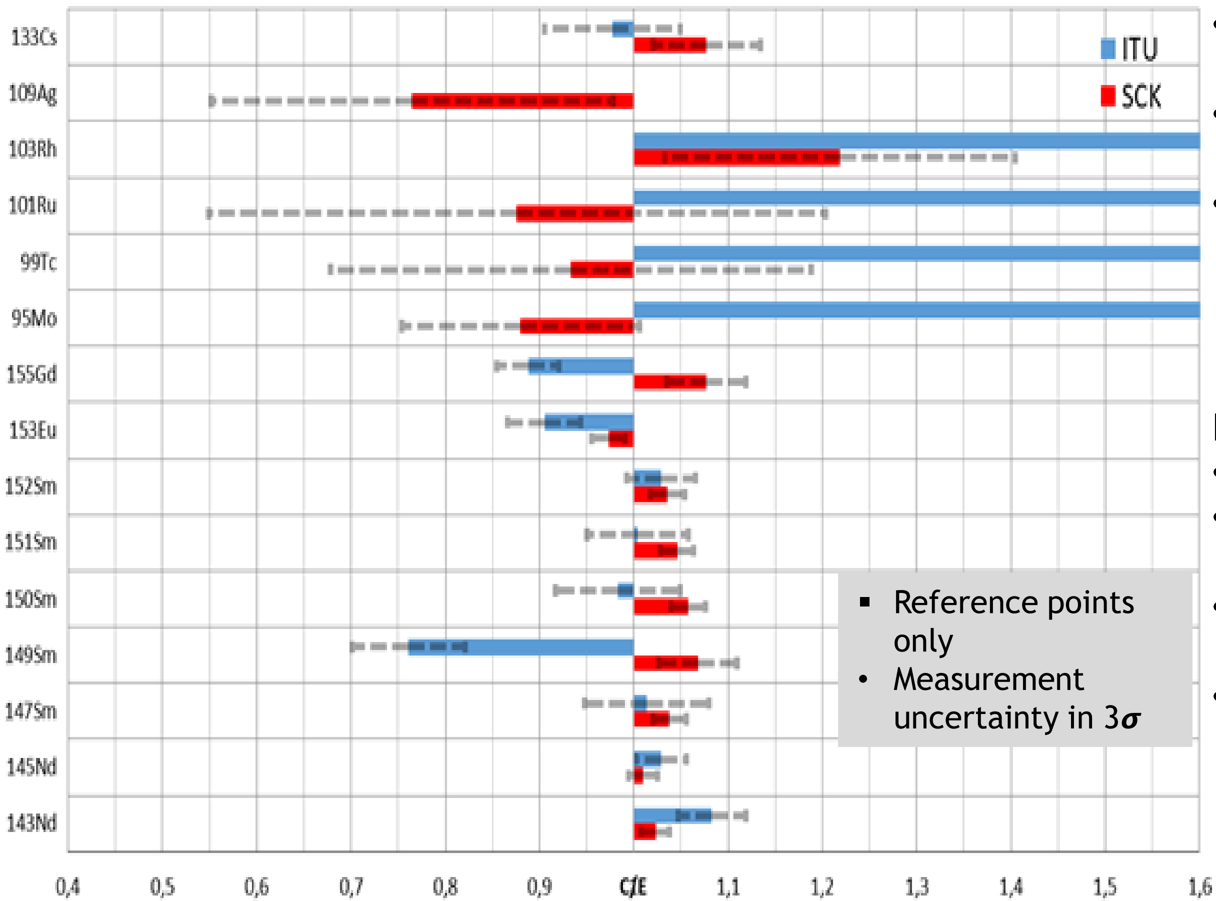
Laboratory	ITU		SCK-CEN	
	Normalization factor	Burn up [MWd.kgHM^{-1}]	Normalization factor	Burn up [MWd.kgHM^{-1}]
Nd reference	0.9753	49.47	1.0300	52.24
Nd lower limit	0.9222	46.78	1.0142	51.44
Nd upper limit	1.0285	52.17	1.0457	53.04

SCK-CEN: error about 1.5 % at 3σ (leads to a total uncertainty on the BU of ~ 0.8 MWd.kgHM^{-1})

ITU: error of 5-6 % at 3σ (leads to a total uncertainty on the BU of ~3 MWd.kgHM^{-1})

Validation results – Fission products

ENDF/B-VII.1 library



- Reference points only
- Measurement uncertainty in 3σ

SCK-CEN analysis.

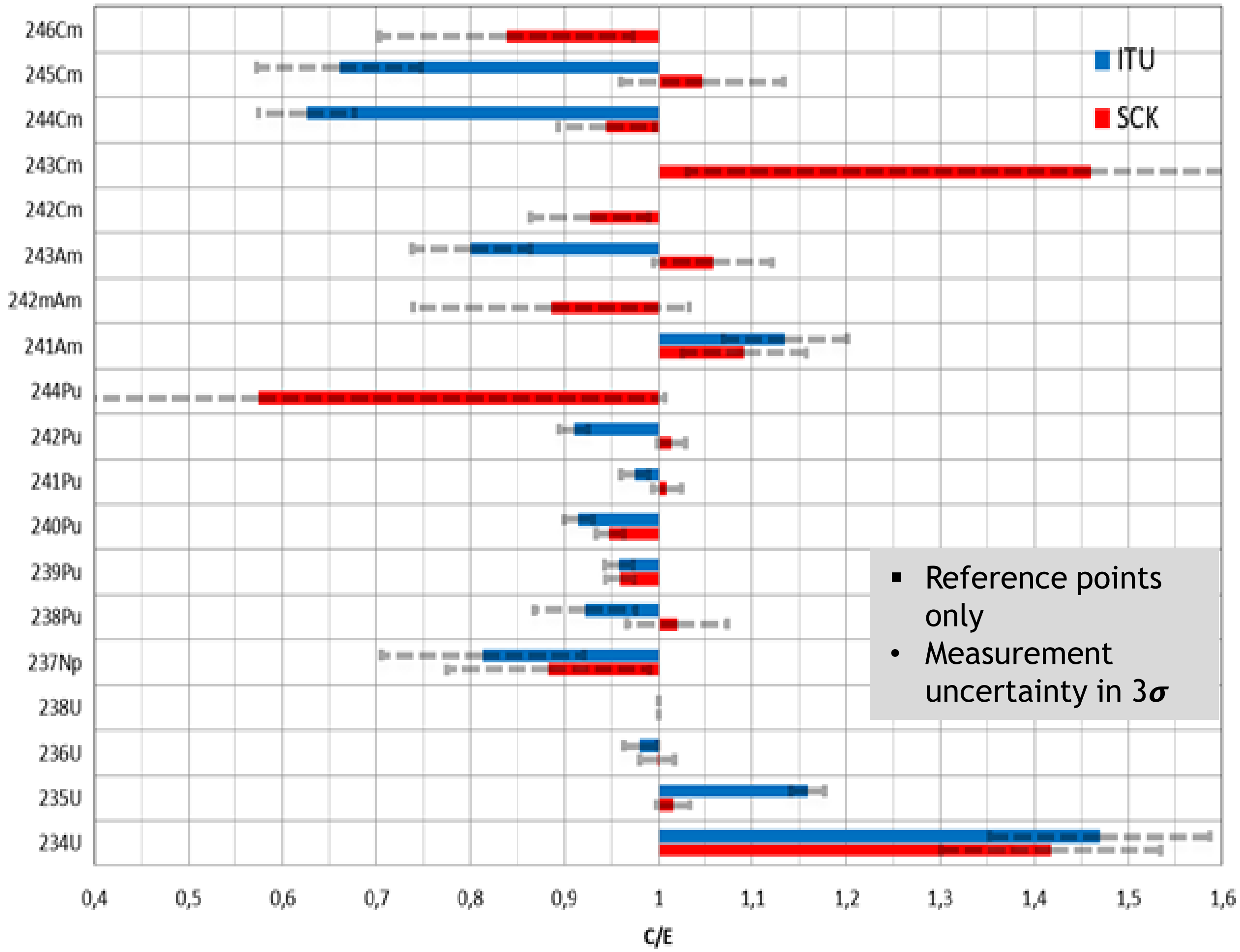
- The Nd content is very well predicted within about 1-2 %
- FP are generally predicted within 20 %.
- Exceptions are: ^{103}Rh and ^{109}Ag (but large measurement uncertainties).

ITU analysis:

- The Nd content is within 5-10 %.
- The other FP are generally predicted within 20 %.
- Exceptions are: ^{149}Sm, ^{103}Rh, ^{95}Mo, and ^{99}Tc.
- ^{149}Sm and ^{155}Gd exhibit a large underestimation, whereas a good agreement in +10 % is obtained for SCK.

Validation results - Actinides

ENDF/B-VII.1 library



- Reference points only
- Measurement uncertainty in 3σ

For the SCK analysis:

- The U and Pu: correctly estimated within 5 % (with the exception of ^{234}U and ^{244}Pu)
- Am: generally overestimated between 1 to 10 %. ^{242m}Am is underestimated by 10 %.
- Cm: generally predicted in 5 to 20 %, ^{243}Cm is overestimated by 50 %.

For the ITU analysis:

- ^{235}U: overestimated by 18 %.
- Pu content: underestimated by 2.5 to 12.5 %.
- Am: within 10 to 20 %
- Cm is largely underestimated.

Impact ENDF/B-VIII.0 vs ENDF/B-VII.1

Actinides				Fission products		
U234	0.73	%		Mo95	-0.08	%
U235	3.36	%		Tc99	0.17	%
U236	-1.57	%		Ru101	0.07	%
U238	-0.01	%		Rh103	0.38	%
Np237	-1.62	%		Ag109	1.11	%
Pu238	-1.27	%		Cs133	0.25	%
Pu239	-0.21	%		Nd143	0.03	%
Pu240	-0.76	%		Nd145	0.04	%
Pu241	0.41	%		Sm147	0.58	%
Pu242	1.13	%		Sm149	0.45	%
Pu244	-5.60	%		Sm150	0.36	%
Am241	0.51	%		Sm151	0.55	%
Am242M	0.80	%		Sm152	0.91	%
Am243	0.73	%		Eu153	-0.47	%
Cm242	0.70	%		Gd155	-0.91	%
Cm243	0.61	%				
Cm244	0.14	%				
Cm245	-0.96	%				
Cm246	-0.79	%				

At end of irradiation time

Very low discrepancies on both actinides and fission products

Maximum discrepancies:

- ^{244}Pu decrease of -5.6 %
- ^{235}U increase of 3.4 % with ENDF/B-VIII.0.

Impact ENDF/B-VIII.0 + IRSN $H(H_2O)$ vs ENDF/B-VIII.0

Actinides				Fission products		
U234	0.50	%		Mo95	0.00	%
U235	-0.10	%		Tc99	0.07	%
U236	0.07	%		Ru101	-0.01	%
U238	0.00	%		Rh103	-0.11	%
Np237	-0.59	%		Ag109	0.39	%
Pu238	-0.30	%		Cs133	0.09	%
Pu239	-0.45	%		Nd143	-0.04	%
Pu240	-0.34	%		Nd145	0.02	%
Pu241	-0.10	%		Sm147	0.34	%
Pu242	0.24	%		Sm149	-0.10	%
Pu244	0.13	%		Sm150	-0.06	%
Am241	-0.13	%		Sm151	0.03	%
Am242M	-0.01	%		Sm152	0.36	%
Am243	0.14	%		Eu153	-0.10	%
Cm242	0.17	%		Gd155	-0.70	%
Cm243	0.41	%				
Cm244	0.23	%				
Cm245	-0.53	%				
Cm246	-0.40	%				

At end of irradiation time

Very low discrepancies on both actinides and fission products

Maximum discrepancies:
^{237}Np decrease of -0.6 % with new IRSN $H(H_2O)$ TSL.

Conclusions

- VESTA 2.2.0 depletion calculations were performed using different libraries and $H(H_2O)$ TSL data for the ARIANE.GU3 sample extracted from a PWR UO_2 fuel rod of the SFCOMPO database.
    - The recent **$H(H_2O)$ TSL at 600 K in ENDF/B-VIII.0**, compared to TSL from JEFF **does not show any significant impact.** Moreover, adding the new ENDF/B-VIII.0 UO_2 TSL at 900 K does not show neither any significant impact.
    - The **new IRSN $H(H_2O)$ TSL at 600 K show a slight over-estimation of roughly 30-50 pcm** during irradiation, compared to full ENDF/B-VIII.0.

- **VESTA 2.2.0 validation results on the ARIANE.GU3 fuel sample using ENDF/B-VII.1 show good agreement with experimental values**, the U and Pu content being generally nicely estimated within 5 % (for the SCK-CEN analysis).

- The **ENDF/B-VIII.0 concentration results agree pretty well with the ENDF/B-VII.1 ones**, which **is also the case when using the new IRSN $H(H_2O)$ TSL at 600 K within ENDF/B-VIII.0**.

Thanks!

Any question ?

Back up

VESTA 2.1.5 validation database

- Experimental validation of VESTA 2.1.5
  - MCNPX 2.6.0 & PHOENIX
  - JEFF 3.1 & ENDF/B-VII.0 nuclear data

- The experimental validation database for VESTA : 96 cases
  - 42 samples with radiochemical analysis of actinides and fission products : for 65 nuclides (versus 53 for VESTA 2.0.0)
  - 54 complete fuel assemblies with decay heat measurements

Program	Reactor	Fuel	Type	Samples
ARIANE	PWR	UO_2 & MOX	Chemical assay	6
MALIBU	PWR	UO_2 & MOX	Chemical assay	5
REBUS	PWR	UO_2	Chemical assay	1
ISTC2670	VVER	UO_2	Chemical assay	8
JAERI	PWR	UO_2 & Gd_2O_3-UO_2	Chemical assay	22
CLAB	PWR	UO_2	Decay heat	34
GE-MORRIS	PWR	UO_2	Decay heat	14
HEDL	PWR	UO_2	Decay heat	6
Total				**96**

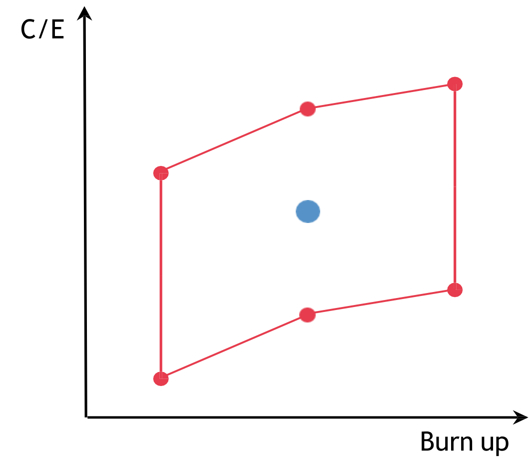
VESTA validation procedure

- Estimate the sample BU and provide a reference C/E value for each nuclide
- An iterative procedure:
  - As long as the C/E of [$^{145}Nd+^{146}Nd+^{148}Nd$] is different from 1, we modify the power (we scale the irradiation history)
  - For every iteration, determine the C/E value and its error for the burn up tracers

    [redacted] [redacted]

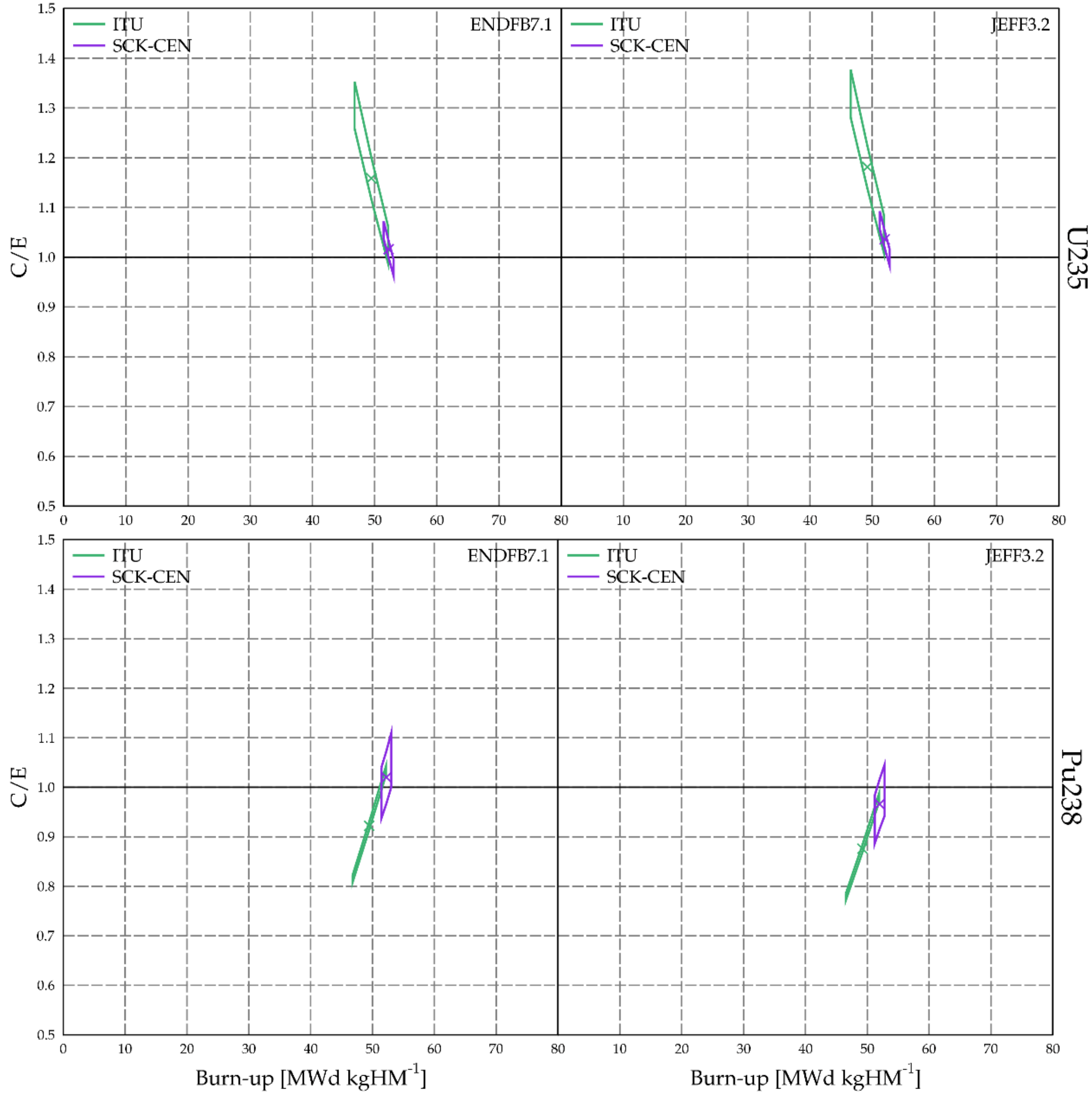
  - Estimate a new scaling factor

    [redacted]



- ^{238}U normalization:
  - Transform into a unit that can be derived from the output
  - As the C/E of ^{238}U is ~1, we divide all the C/E results by the C/E of ^{238}U

C/E results



TSL Theoretical formalism

- The double differential cross-section for neutrons with incident energy **E**, secondary energy **E'**, μ is the neutron scattering cosine and scattering angle Ω is related to S(α,β) by :

$$\frac{d^2\sigma}{d\Omega dE'}(E \rightarrow E', \Omega) = \frac{\sigma_b}{4\pi k_B T}\sqrt{\frac{E'}{E}}S(\alpha,\beta)$$

S(α, β)

Thermal Scattering Law

$$\alpha = \frac{E + E' - 2\mu\sqrt{EE'}}{Ak_BT}$$

Momentum transfer

$$\beta = \frac{E' - E}{k_BT}$$

Energy transfer

$$S(\alpha,\beta) = \frac{1}{2\pi}\int_{-\infty}^{+\infty} e^{\iota\beta\hat{t}}\, e^{-\gamma(\hat{t})} d\hat{t}$$

$$\gamma(\hat{t}) = \alpha\int_{-\infty}^{+\infty} P(\beta)(1 - \, e^{-\iota\beta\hat{t}})\, e^{-\beta/2} d\beta$$

$$P(\beta) = \frac{\boxed{\rho(\beta)}}{2\beta\sinh(\beta/2)}$$

Frequency spectrum / Density of states

V. Jaiswal thesis, 2018

MOLECULAR DYNAMICS SIMULATIONS

↓

- MD is the solution of the classical equations of motion for atoms and molecules to obtain the time evolution of the system.

↓

$$m_i \frac{\partial^2 \boldsymbol{r_i}}{\partial t^2} = \boldsymbol{F}_i \quad , i=1\ldots.N$$

- The forces are the negative derivatives of a potential function

$$\boldsymbol{U}\,(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_N)$$

Potential/Force field

$$\boldsymbol{F_i} = -\frac{\partial \boldsymbol{U}}{\partial \boldsymbol{r_i}}$$

- For any given configuration one can calculate the acceleration of each particle and hence the velocity and the position of the atoms.

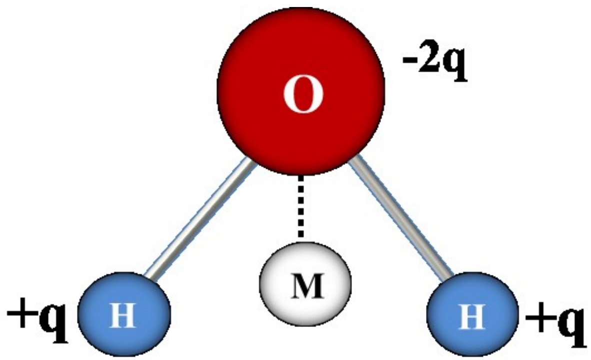
↓

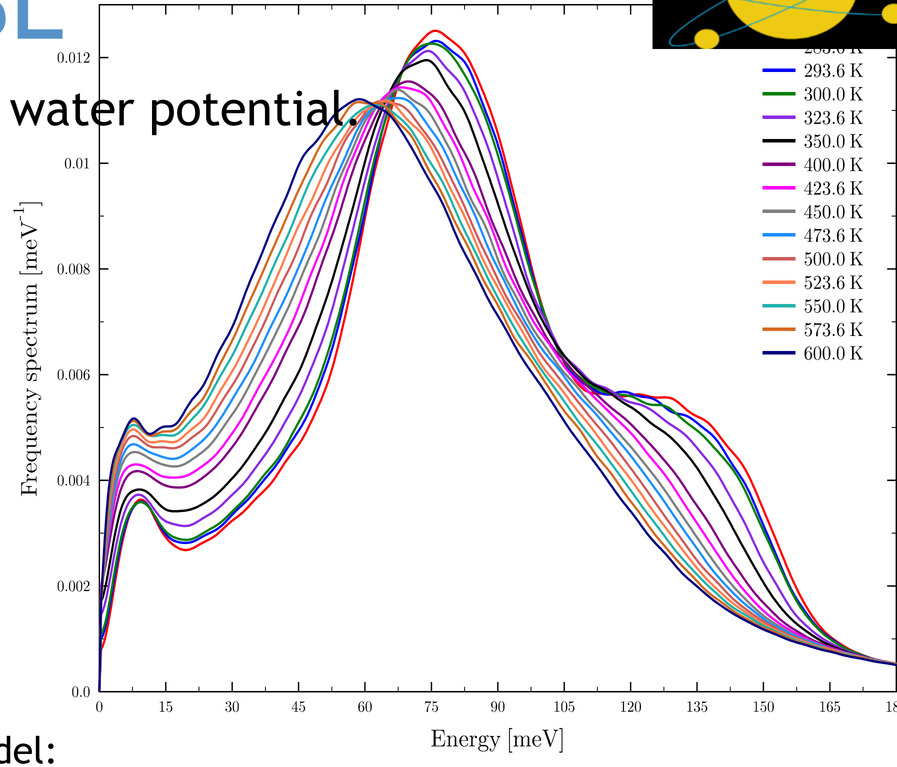
TRAJECTORIES

V. Jaiswal thesis, 2018

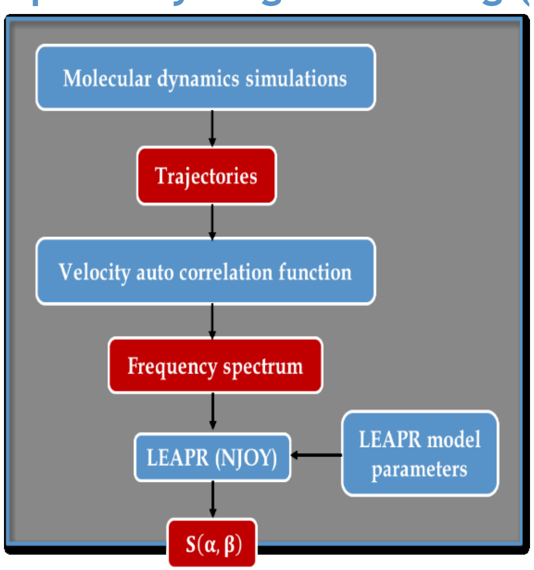
New IRSN $H(H_2O)$ TSL

PolarisMD code and TCPE polarizable rigid water potential.





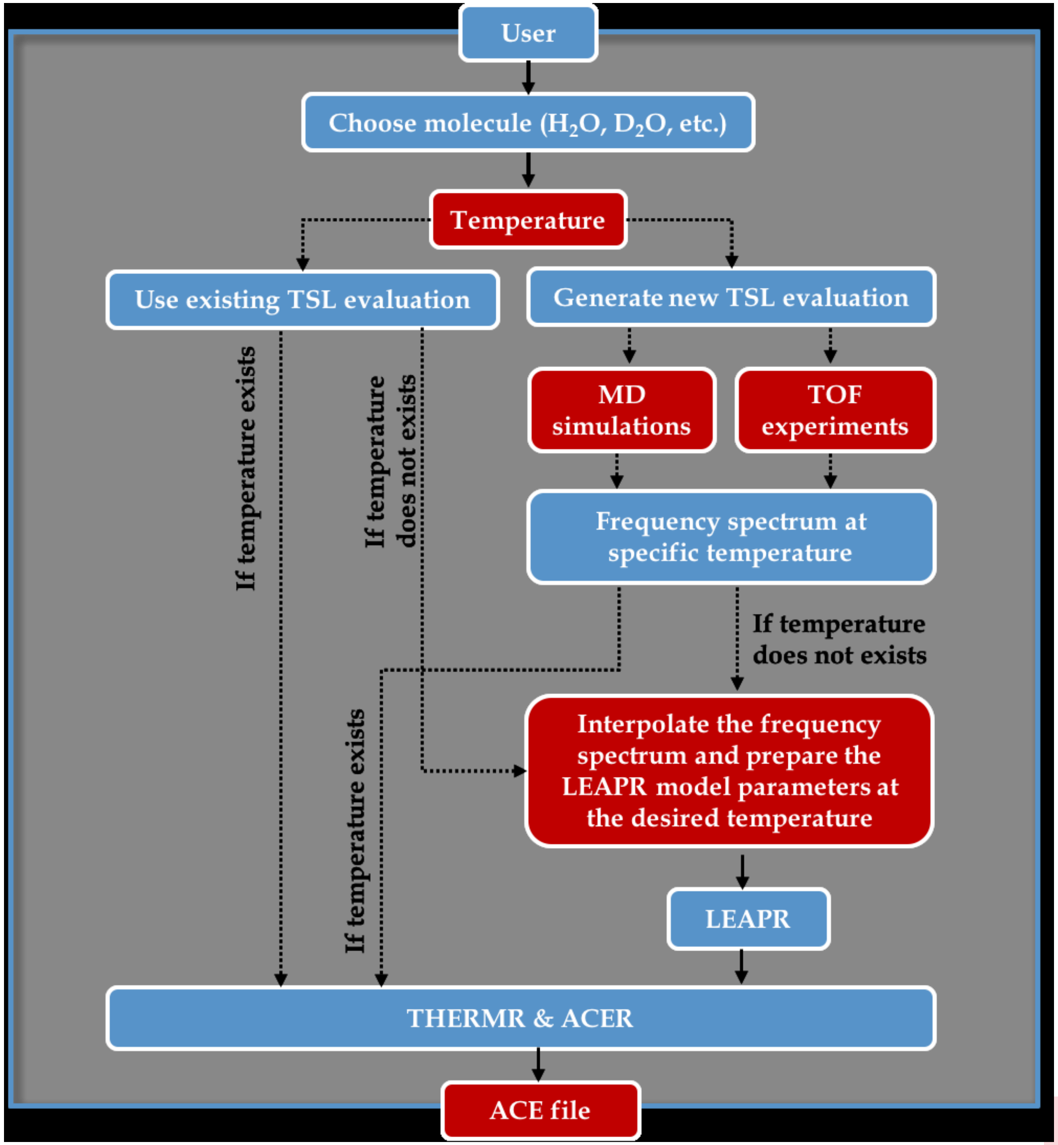
Rigid polarizable TCPE water model with explicit hydrogen bonding (This work)



- Polarizable model:
  - respond to fluctuations in the electric field due to molecular motion.
  - is a promising solution for reactor physics applications at high temperatures and pressure.
- TCPE water model accurately reproduces the experimental self-diffusion coefficient of water for a series of high temperatures.

V. Jaiswal thesis, 2018

New IRSN $H(H_2O)$ TSL



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