Validation of Thermal Scattering Laws for Light Water at Elevated Temperatures with Diffusion Experiments

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Motivation

- Light water (H-H_2O) is the most important ENDF-format thermal neutron scattering law (TSL) material for many reactor applications.

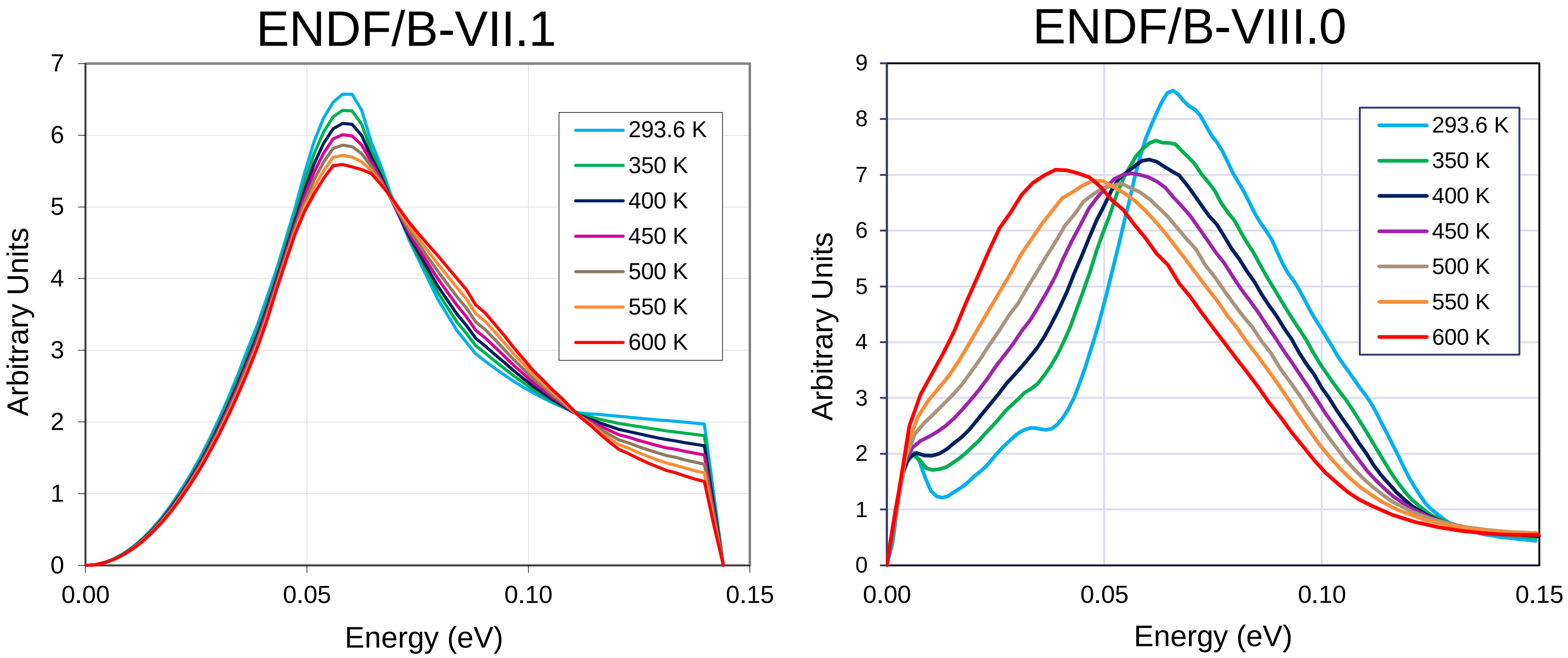
- Many reactor applications involve elevated temperature (T) and pressure (P).

- Direct validation of H-H_2O TSL performance for these conditions is limited.

- Early efforts (1950s and 1960s) to characterize thermal neutron scattering in water for reactor applications produced more than 100 publications of experimental measurements of neutron diffusion over a range of elevated T.

Temperature-Dependent H-H_2O Phonon Spectra

Phonon and diffusion information is independently defined for each *T*



Mattes and Keinert IKE model (2005) based on **linear interpolation** of DDXS measurements at 295 K and 550 K, Page and Haywood, AERE Report 5578, United Kingdom (1968).

CAB molecular dynamics simulation model by Márquez Damián et al., *Annals of Nuclear Energy* **65** (2014).

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Importance of Temperature-Dependent Validation

- The dynamics of H_2O molecules in liquid water change significantly with respect to *T*, along with density, and these changes are non-linear.

- Consequently, thermal neutron scattering microscopic cross sections, and probabilities in energy and angle, vary non-linearly with *T*.

- The accuracy of calculated thermal scattering cross sections depends on the temperature-dependent phonon and diffusion information in the H-H_2O TSL.

- Unlike solids and “fast” evaluations, validation of H-H_2O TSL performance at one *T* provides little direct assurance of acceptable performance at another.

THEORETICAL BASIS FOR USING NEUTRON DIFFUSION MEASUREMENTS FOR TSL VALIDATION

Linking Diffusion Theory to Experiment (Time-Dependent Case)

Time-dependent one-speed diffusion equation for a homogeneous medium:

$$\frac{1}{v}\frac{\partial\varphi(\boldsymbol{r},t)}{\partial t} - D\nabla^2\varphi(\boldsymbol{r},t) + \Sigma_\mathrm{a} = S(\boldsymbol{r},t)$$

Fundamental-mode solution for a non-multiplying system with no neutron source:

$\varphi(\boldsymbol{r},t) = \varphi_0(\boldsymbol{r})e^{-\alpha t}$, where

$$\alpha = v\Sigma_\mathrm{a} + vD{B_\mathrm{g}}^2$$

The blue expression for α is from one-speed diffusion theory, where ${B_\mathrm{g}}^2$ is geometric buckling.

- For a real homogeneous source-free non-multiplying system with a thermalized neutron flux, the solution form in red is physical.
- The physical time eigenvalue α is independent of diffusion theory. Using the pulsed-neutron die-away (PNDA) method, it may be measured or calculated by simulation.

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Linking Diffusion Theory to Experiment (Time-Dependent Case)

The physical α can be expanded as as a Taylor series in ${B_\mathrm{g}}^2$:

$$\alpha = \overline{v\Sigma_\mathrm{a}} + \overline{vD}{B_\mathrm{g}}^2 - C{B_\mathrm{g}}^4 + O\left({B_\mathrm{g}}^6\right) \approx a_\mathrm{eff} + D_\mathrm{eff}{B_\mathrm{g}}^2 - C{B_\mathrm{g}}^4$$

Now, a_eff is an “effective” absorption term, D_eff is an “effective” diffusion coefficient, and the one-speed approximation is relaxed (a_eff and D_eff are spectrally averaged).

- The parameter C is a diffusion cooling coefficient associated with preferential “transport” and boundary leakage of higher-energy neutrons in the thermal spectrum.
- The second-order and higher terms in buckling are essentially “transport corrections” to the diffusion-theory time-eigenvalue form $\alpha = v\Sigma_\mathrm{a} + vD{B_\mathrm{g}}^2$.
- By determining α over a range of ${B_\mathrm{g}}^2$ (or for samples of varying dimensions), a_eff, D_eff, and C can be calculated with a quadratic (or higher order) fit of α to ${B_\mathrm{g}}^2$.

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Linking Diffusion Theory to Experiment (Static Case)

If an external source makes up for losses in the flux such that it is constant in time, the one-speed diffusion equation for a homogeneous medium in this stationary case is:

$$\nabla^2\varphi(\boldsymbol{r}) - \frac{1}{L^2}\varphi(\boldsymbol{r}) = -\frac{S(\boldsymbol{r})}{D},$$

where L (the neutron diffusion length) $\equiv \sqrt{D/\Sigma_{\mathrm{a}}} = \frac{1}{\sqrt{6}} * \text{RMS distance to absorption}$

For an infinite homogeneous non-multiplying system with a point source at $r = 0$, the fundamental-mode flux profile solution converges as $r \rightarrow \infty$ to the form:

$$\varphi(r) = \varphi_0\frac{e^{-\kappa r}}{r}, \text{ where } \kappa = 1/L$$

For a real system, the form in red still describes the physical asymptotic solution far from a source or boundary, where the spatial eigenvalue κ depends on the physical L.

Equivalence of the Time-Dependent and Static Cases

With $L \equiv \sqrt{D/\Sigma_\mathrm{a}}$, the one-speed static case $\alpha = v\Sigma_\mathrm{a} + vD{B_\mathrm{g}}^2 = 0$ requires ${B_\mathrm{g}}^2 = -1/L^2$, since v, Σ_a, and D are physical properties that cannot be negative.

${B_\mathrm{g}}^2$ characterizes the boundary leakage effect on the flux spatial curvature and decay rate.

Requiring negative buckling (${B_\mathrm{g}}^2 = -1/L^2$) to yield $\alpha = 0$ is equivalent to stating that net leakage is negative due to the presence of an external source.

$\alpha \approx a_\mathrm{eff} + D_\mathrm{eff}{B_\mathrm{g}}^2 - C{B_\mathrm{g}}^4$ is a more physical representation of α.
Solving the quadratic expansion of $\alpha \approx a_\mathrm{eff} + D_\mathrm{eff}{B_\mathrm{g}}^2 - C{B_\mathrm{g}}^4$ with ${B_\mathrm{g}}^2 = -1/L^2$ yields:

$$L = \sqrt{\frac{D_\mathrm{eff}}{2a_\mathrm{eff}}\left(1 + \sqrt{1 + \frac{4a_\mathrm{eff}C}{D_\mathrm{eff}^2}}\right)} \approx \sqrt{\frac{D_\mathrm{eff}}{a_\mathrm{eff}} + \frac{C}{D_\mathrm{eff}}} \approx \sqrt{\frac{D_\mathrm{eff}}{a_\mathrm{eff}}}$$

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Equivalence of the Time-Dependent and Static Cases

The solution $L = \sqrt{\frac{D_{\mathrm{eff}}}{2a_{\mathrm{eff}}}\left(1 + \sqrt{1 + \frac{4a_{\mathrm{eff}}C}{D_{\mathrm{eff}}^2}}\right)}$ was derived from the static case in terms of the expanded diffusion coefficients from the time-dependent case.

However, the thermal neutron diffusion length L is an intrinsic property of a material.

- L physically depends on the density, temperature, absorption, and integral and differential thermal scattering properties of the material.
- L can be calculated equivalently as $L = 1/\kappa$ by determining the spatial eigenvalue κ (with a fixed source) or by the PNDA method using the equation form for L above.
- The neglect of higher-order coefficients in the equation for L above has virtually no impact for light water (due to weak absorption and near-linear angular dependence).

Physical Geometric Buckling

- The form ${B_g}^2 = \left(\frac{\pi}{R+z_0}\right)^2$ is given by diffusion theory for spherical geometry, where the extrapolation distance z_0 serves as a vacuum boundary correction.

- The physical treatment of z_0 and ${B_g}^2$ in PNDA experiments have been the subject of considerable study, including for light water and for spherical systems.

- While z_0 and ${B_g}^2$ are artifacts of diffusion theory, for real systems of arbitrary geometry, the physically appropriate ${B_g}^2$ have values which reproduce the physical time eigenvalues $\alpha = a_{\mathrm{eff}} + D_{\mathrm{eff}}{B_g}^2 - C{B_g}^4$ and yield L consistent with the physical spatial eigenvalue $\kappa = 1/L$.

- The appropriate z_0 is a function of material density, material scattering and absorption properties, and geometry. This relationship is given by $z_0 = \lambda_{\mathrm{tr}}F_g$. The geometry factor F_g is shape-dependent and is a nearly-constant function of ${B_g}^2$, except for very small systems.

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DEVELOPING AN EMPIRICAL RELATIONSHIP OF THE NEUTRON DIFFUSION LENGTH AS A FUNCTION OF TEMPERATURE

Incorporation of Historical Data

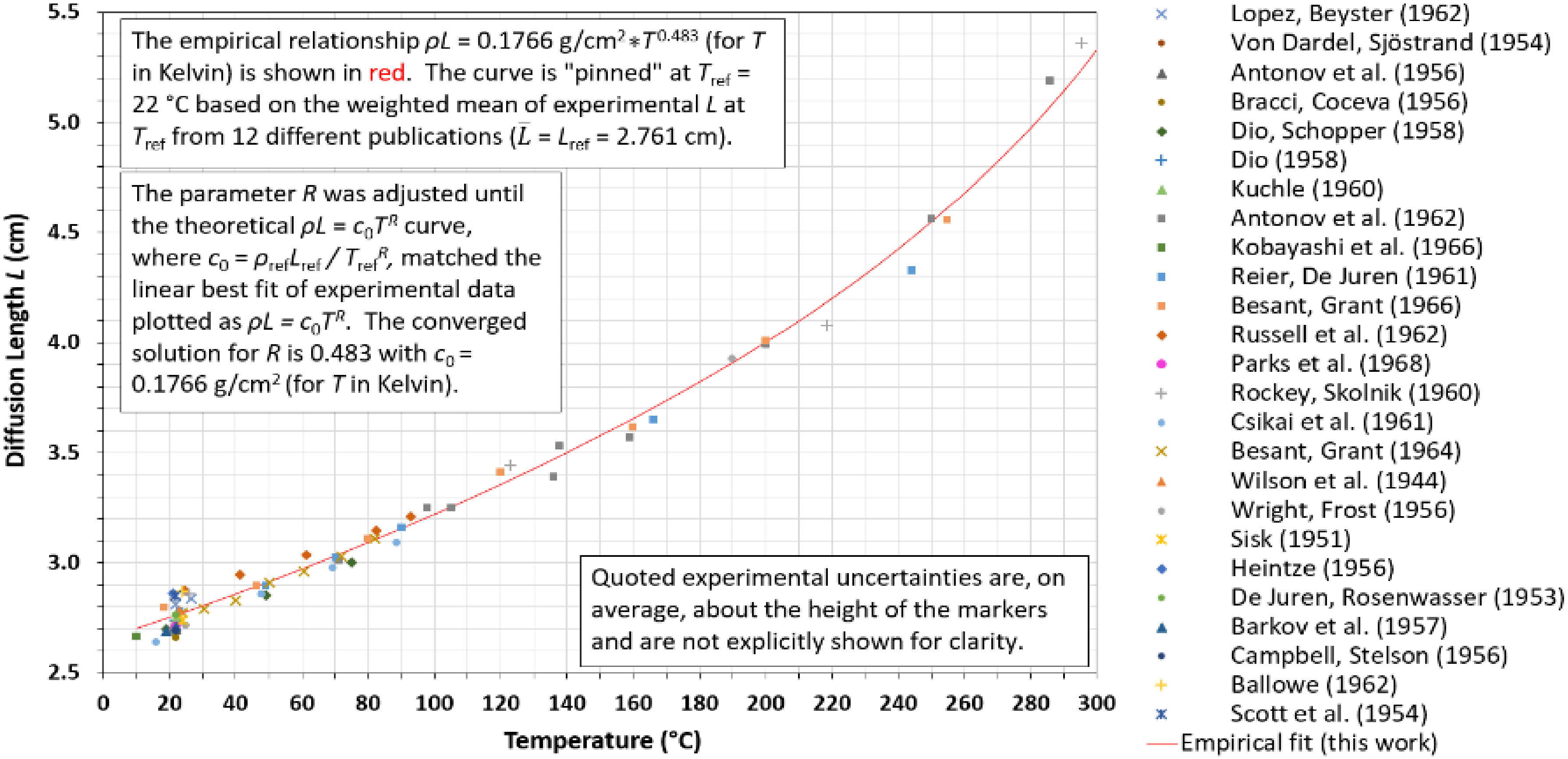
- The physical variation of L with T depends on the non-linear relationship of density (ρ) vs. T as well as on changes in the dynamics of H_2O molecules (such as changes in clustering and rotational hindrance).

- Under the assumption that microscopic cross sections are T-independent and have $1/v$ dependence over a Maxwell-Boltzmann neutron spectrum, diffusion theory predicts that ρL should vary as $\sqrt{T}$.

- Physically, experimental data has been shown to follow the form $\rho L = T^R$, where R is slightly less than 0.5.

- In the present work, experimental L data at 49 reported temperatures from 25 publications (using several types of static and time-dependent methods) is compiled.

Experimental L Data and Empirical Temperature Fit



The empirical relationship ρL = 0.1766 g/cm^2∗$T^{0.483}$ (for T in Kelvin) is shown in red. The curve is "pinned" at T_{ref} = 22 °C based on the weighted mean of experimental L at T_{ref} from 12 different publications ($\bar{L}$ = L_{ref} = 2.761 cm).

The parameter R was adjusted until the theoretical $\rho L = c_0 T^R$ curve, where $c_0 = \rho_{ref} L_{ref} / T_{ref}^R$, matched the linear best fit of experimental data plotted as $\rho L = c_0 T^R$. The converged solution for R is 0.483 with c_0 = 0.1766 g/cm^2 (for T in Kelvin).

Quoted experimental uncertainties are, on average, about the height of the markers and are not explicitly shown for clarity.

$$\rho L = (0.1766\ \mathrm{g/cm^2}) T^{0.483} \text{ (for } T \text{ in Kelvin)}$$

$$L_{\mathrm{ref}}\ (\text{at } T_{\mathrm{ref}} = 22\ \text{°C}) = 2.761 \pm 0.009\ \mathrm{cm}$$

Uncertainties and Biases

Clear systematic biases exist between publications, where the data spread exceeds what may be reasonably expected based on the authors' quoted uncertainties.

L data from 10 PNDA sources:

0.8% below empirical curve

L data from 15 static sources:

0.2% above empirical curve (with half the quoted uncertainty of PNDA)

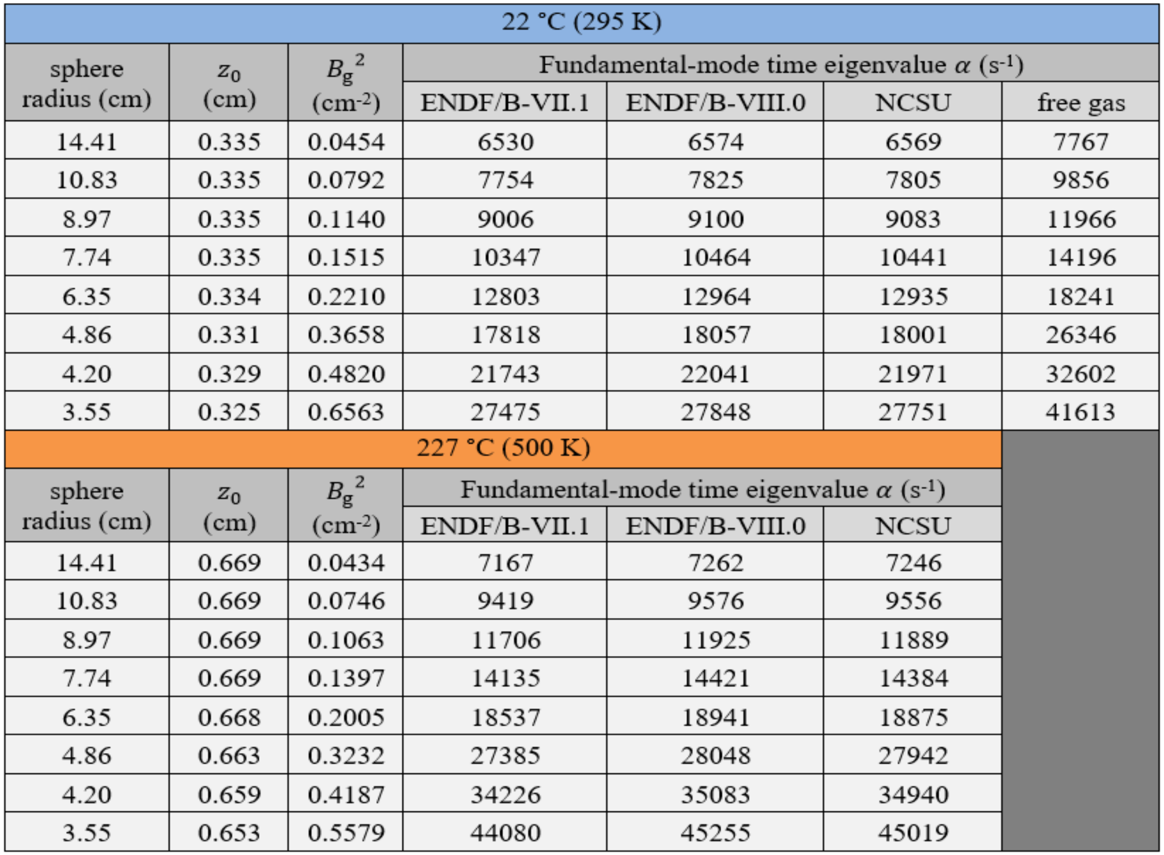
Taking into account the ambiguity of the observed systematic biases, a best estimate of 0.5% is given for the uncertainty in calculating L with the empirical function $\rho L = (0.1766\ \mathrm{g/cm^2})T^{0.483}$ (for T in Kelvin).

RESULTS OF

PNDA SIMULATION OF H-H2O TSLs

WITH MC21

PNDA Model Data and Time Eigenvalue Results



22 °C (295 K)

sphere radius (cm)	z_0 (cm)	B_g^2 (cm^{-2})	Fundamental-mode time eigenvalue α (s^{-1})			
			ENDF/B-VII.1	ENDF/B-VIII.0	NCSU	free gas
14.41	0.335	0.0454	6530	6574	6569	7767
10.83	0.335	0.0792	7754	7825	7805	9856
8.97	0.335	0.1140	9006	9100	9083	11966
7.74	0.335	0.1515	10347	10464	10441	14196
6.35	0.334	0.2210	12803	12964	12935	18241
4.86	0.331	0.3658	17818	18057	18001	26346
4.20	0.329	0.4820	21743	22041	21971	32602
3.55	0.325	0.6563	27475	27848	27751	41613

227 °C (500 K)

sphere radius (cm)	z_0 (cm)	B_g^2 (cm^{-2})	Fundamental-mode time eigenvalue α (s^{-1})		
			ENDF/B-VII.1	ENDF/B-VIII.0	NCSU
14.41	0.669	0.0434	7167	7262	7246
10.83	0.669	0.0746	9419	9576	9556
8.97	0.669	0.1063	11706	11925	11889
7.74	0.669	0.1397	14135	14421	14384
6.35	0.668	0.2005	18537	18941	18875
4.86	0.663	0.3232	27385	28048	27942
4.20	0.659	0.4187	34226	35083	34940
3.55	0.653	0.5579	44080	45255	45019

Room temperature z_0 data from Schmidt and Gelbard (1965).

NCSU TSL is under development.

Temperature Correction of z_0

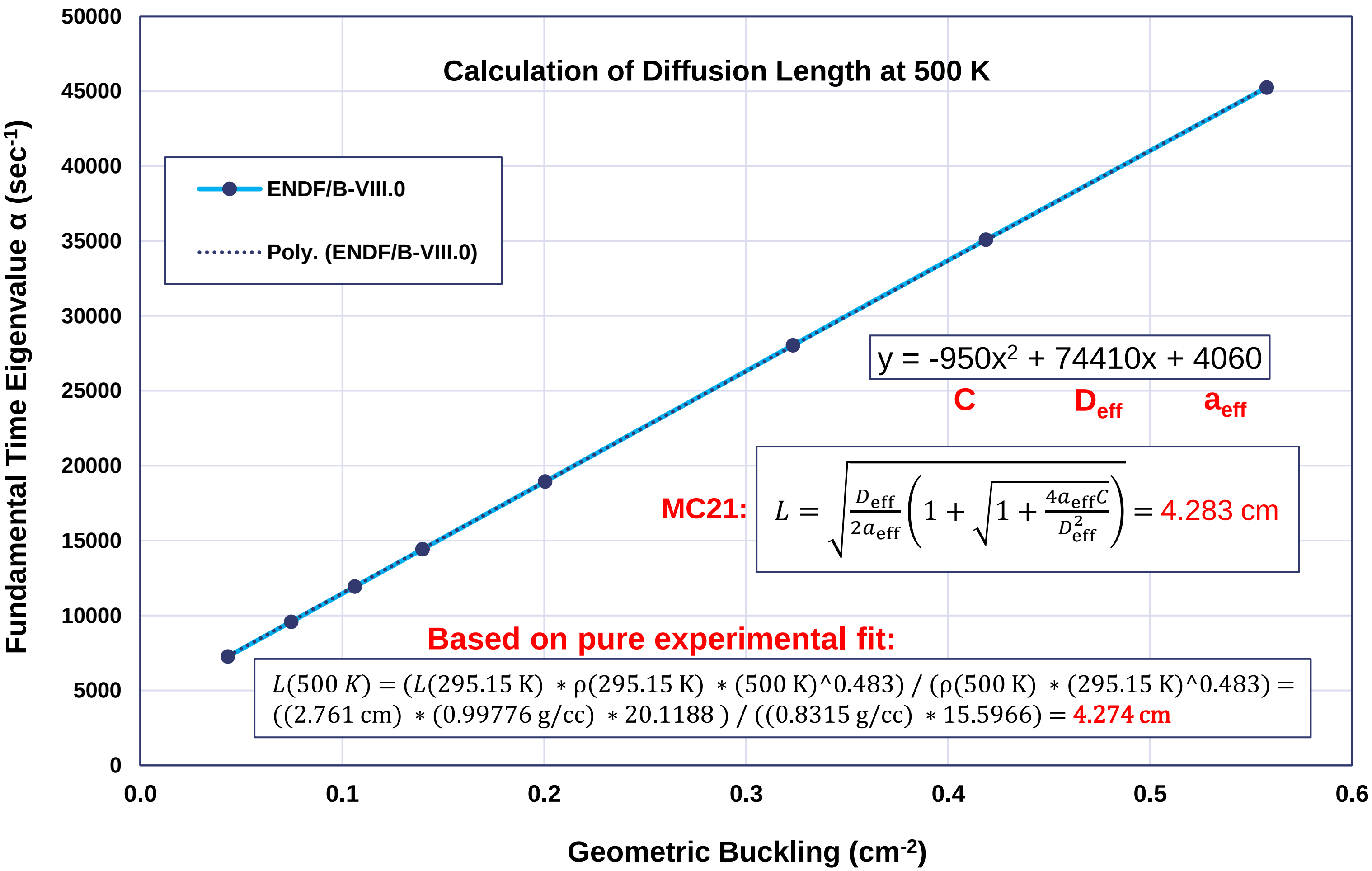
- To determine appropriate z_0 at 500 K, we note that the theoretical value for $L_{500\ \mathrm{K}}$ is given by $L_{500\ \mathrm{K}} = (0.1766\ \mathrm{g/cm^2})(500)^{0.483}/(\rho_{500\ \mathrm{K}} = 0.8315\ \mathrm{g/cm^3})$ = 4.274 cm.

- Using $L \approx \sqrt{D_\mathrm{eff}/a_\mathrm{eff}}$, we can predict $D_{500\ \mathrm{K}}/D_\mathrm{ref} = (\rho_{500\ \mathrm{K}}/\rho_\mathrm{ref})(L_{500\ \mathrm{K}}/L_\mathrm{ref})^2$ = 1.997.

- Given $\lambda_\mathrm{tr} \propto D_\mathrm{eff}$ and $z_0 \propto \lambda_\mathrm{tr}$, the z_0 values at T_ref are first scaled by a factor of 1.997.

- Since ${B_\mathrm{g}}^2$ decreases as T increases, fine adjustments are then made to z_0 to account for the small variation of F_g with ${B_\mathrm{g}}^2$.

MC21 ENDF/B-VIII.0 H-H_2O TSL vs. Empirical at 500 K



$y = -950x^2 + 74410x + 4060$

C **D_{eff}** **a_{eff}**

MC21:

$$L = \sqrt{\frac{D_{\text{eff}}}{2a_{\text{eff}}}\left(1+\sqrt{1+\frac{4a_{\text{eff}}C}{D_{\text{eff}}^2}}\right)} = 4.283\ \text{cm}$$

Based on pure experimental fit:

$L(500\ K) = (L(295.15\ \text{K}) * \rho(295.15\ \text{K}) * (500\ \text{K})\text{^}0.483) / (\rho(500\ \text{K}) * (295.15\ \text{K})\text{^}0.483) =$
$((2.761\ \text{cm}) * (0.99776\ \text{g/cc}) * 20.1188\ ) / ((0.8315\ \text{g/cc}) * 15.5966) =$ **4.274 cm**

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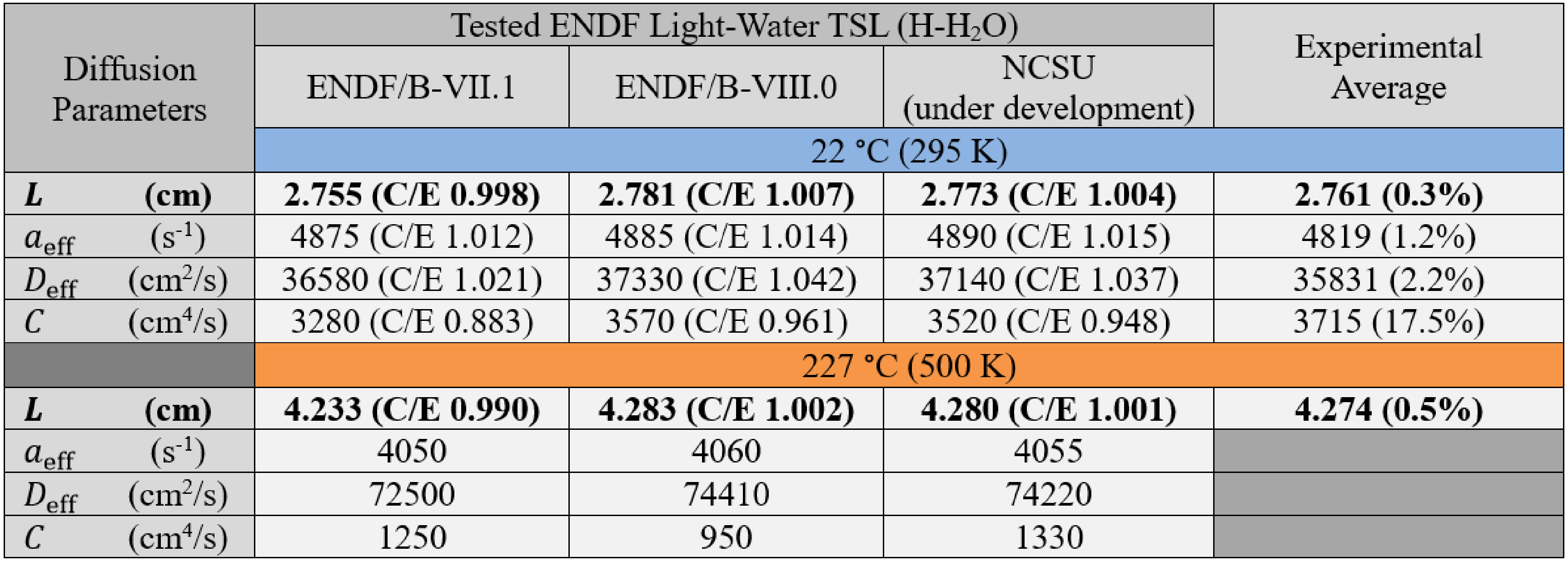
Diffusion Parameter Results Compared to Experimental Averages



Diffusion Parameters	Tested ENDF Light-Water TSL (H-H_2O)			Experimental Average
	ENDF/B-VII.1	ENDF/B-VIII.0	NCSU (under development)	
	22 °C (295 K)			
L (cm)	**2.755 (C/E 0.998)**	**2.781 (C/E 1.007)**	**2.773 (C/E 1.004)**	**2.761 (0.3%)**
a_{eff} (s^{-1})	4875 (C/E 1.012)	4885 (C/E 1.014)	4890 (C/E 1.015)	4819 (1.2%)
D_{eff} (cm^2/s)	36580 (C/E 1.021)	37330 (C/E 1.042)	37140 (C/E 1.037)	35831 (2.2%)
C (cm^4/s)	3280 (C/E 0.883)	3570 (C/E 0.961)	3520 (C/E 0.948)	3715 (17.5%)
	227 °C (500 K)			
L (cm)	**4.233 (C/E 0.990)**	**4.283 (C/E 1.002)**	**4.280 (C/E 1.001)**	**4.274 (0.5%)**
a_{eff} (s^{-1})	4050	4060	4055	
D_{eff} (cm^2/s)	72500	74410	74220	
C (cm^4/s)	1250	950	1330	

Experimental values and uncertainties for a_{eff}, D_{eff}, and C are unweighted and based on available data from 10 publications at or near T_{ref} = 22 °C.

All data for a_{eff} and D_{eff} is adjusted to 22 °C. Data for C is unadjusted. The experimental value and uncertainty for L at 227 °C is based on the empirical fit.

The estimated uncertainty in MC21-calculated L using the given PNDA method is 0.3%.

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Quality of C/E for Expanded Diffusion Coefficients

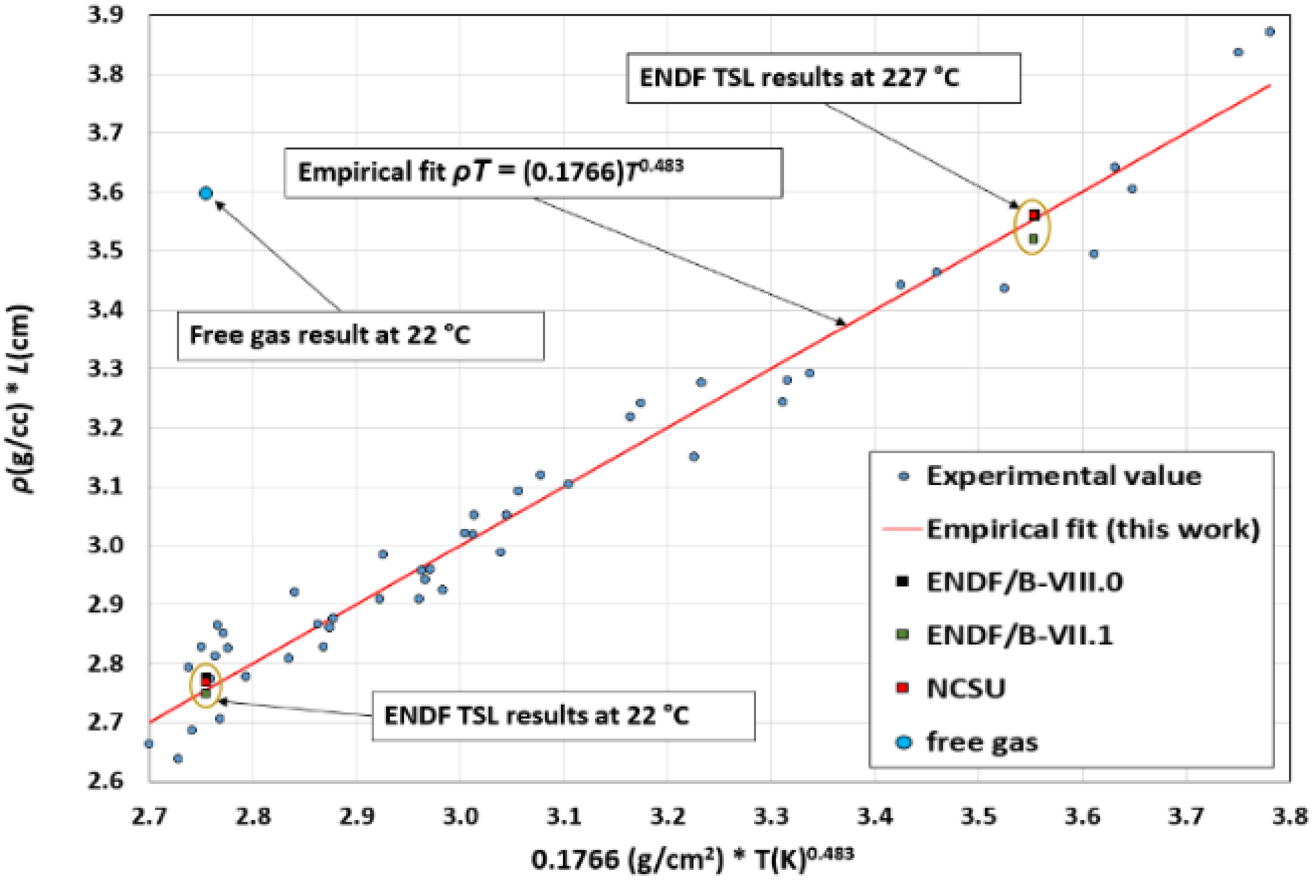
- The experimental a_{eff}, D_{eff}, and C data used in calculating C/E values at 22 °C is primarily from PNDA sources.

- Considering the PNDA-based experimental L data used in the work is biased about 1% lower than the static-method L data, it is possible that the experimental D_{eff} average is biased too low (since $L \approx \sqrt{D_{\mathrm{eff}}/a_{\mathrm{eff}}}$, and a_{eff} can be calculated more accurately than D_{eff}).

- It is notable that the MC21-calculated $D_{500\ \mathrm{K}}/D_{\mathrm{ref}}$, averaged over the two MD-generated TSLs tested (ENDF/B-VIII.0 and NCSU), is 1.996, nearly exactly the value of 1.997 predicted based on the empirical fit derived from experimental L data.

- Uncertainties in C, whether calculated by experiment or Monte Carlo simulation, will always be large. Higher-order diffusion effects are very weak in light water, and effectively probing C requires extremely small geometries that can be impractical.

MC21 Results in Empirical Fit Space



Note that O is always treated as a free gas (as is justified by the results). However, treating H as a free gas (shown above) results in enormous deviation of L from physical reality.

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Conclusions

- The ability to accurately calculate the thermal neutron diffusion length L in light water with MC21 PNDA simulations at very different temperatures has been established by leveraging experimental data from numerous publications.

- The calculated L is consistent with experimental measurements and sufficiently sensitive to different H-H2O TSL physics models to use the method as a TSL integral performance benchmark, if not a validation basis.

- This method allows comparison of calculated L to experiment for light water at arbitrary T.

- The diffusion-based validation method for H-H_2O TSLs has the advantage of being isolated to the physics of the material under consideration. There is no influence of nuclear data uncertainties or effects from other materials.

- Repeating the method over a fine mesh of temperatures should result in a smooth inflectionless curve of L vs. T. Any anomalies in the underlying TSL physics models as a function of T could be easily identified, which is difficult with critical benchmarks.