Characterization of Thermal Conductivity using Deterministic Phonon Transport in Rattlesnake

Jackson R. Harter, P. Alex Greaney, Ph.D, Todd S. Palmer, Ph.D

Radiation Transport Group, Department of Nuclear Engineering and Radiation Health Physics, Oregon State University
Corvallis, OR
harterj@onid.oregonstate.edu, alex.greaney@oregonstate.edu, palmerts@engr.orst.edu

INTRODUCTION

Measuring thermal conductivity in nuclear fuel is an important aspect of the safe operation of nuclear power plants or research reactors. In the past, knowledge of thermal conductivity in nuclear fuel has been obtained primarily through empirical studies and measurements. Studies are conducted with fuel in different stages of burnup. Measurements are taken with respect to the microstructure, determining a thermal conductivity for a specific concentration of fission products.

However, to accurately determine thermal conductivity in nuclear fuel, other factors must be taken into consideration. A first principles, physics-based calculation of thermal conductivity must involve factors such as the microstructure of nuclear fuel, as this parameter constantly changes during the fission process through the formation of isotopic decay products. Oxide nuclear fuel is a ceramic, and heat transport in ceramics is dominated by phonon transport mechanics. Computational materials science has produced a body of work in phonon transport at the nanoscale [1, 2]. Impurities in the bulk material influence the transport of energy at the fundamental level, altering the scattering behavior of phonons and electrons. Conventionally, heat transport follows classical physics based on the heat equation derived from Fourier's law

$$\boldsymbol{q} = -k\boldsymbol{\nabla}T, \tag{1}$$

where k is the thermal conductivity, a second rank tensor. At the nanoscale heat transport behaves differently [1] and consideration of alternative means of identifying thermal conductivity in nuclear fuel is necessary for continued development and safe operation of nuclear facilities.

MOOSE Framework

Idaho National Laboratory (INL) has developed a sophisticated framework for solving coupled partial differential equations, the Multi-physics Object Oriented Simulation Environment (MOOSE) [3]. Within MOOSE are a number of modules which solve the equations for specific physics applications, e.g., neutron transport, thermal hydraulics, materials science, or probabilistic risk assessment. One of these modules, Rattlesnake, is a neutron Boltzmann transport equation (BTE) solver using the Self-Adjoint Angular Flux (SAAF) formulation.

The goal of our research is to construct a module for MOOSE which simulates phonon transport using Rattlesnake as the particle transport engine. We will use phonon transport solutions to bridge the gap between molecular dynamics and engineering scale simulations and better predict thermal conductivity in bulk heterogeneous nuclear fuels with fission product defects (UO_2, PuO_2). Using Rattlesnake, we have

replicated phonon transport results from a bulk homogeneous material phonon transport problem solved via a finite difference approach. We show that the phonon modified BTE can be derived in a second order formulation, and phonon transport can be simulated using this method. We are now in the process of creating new geometries (investigation of size and impurity effects) and conducting 3D transport simulations of heterogeneous media.

METHODS

A one dimensional neutron transport code using the SAAF of the BTE was written based on work by Morel and McGhee [4]. Written in MATLAB [5], the code is a steady-state, monoenergetic, source iteration, S_N transport solver and uses a finite difference discretization scheme for a slab geometry application. The code was adapted to model phonon transport, and successfully replicated test data by Yilbas and Bin Mansoor [2], who used forward and backward differencing schemes to numerically solve the equation of phonon radiative transport (EPRT) [1]. They were successful, but the two schemes were only stable when coefficients were positive for the forward case, and negative for the backward case. With the SAAF approach, we have mitigated some of the challenges they faced. Fig. 1 is a comparison of our results to their numerical simulations.
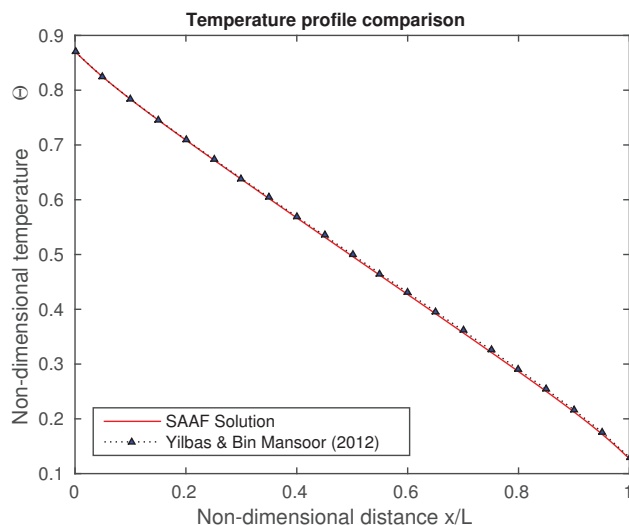

Fig. 1. Results of 1-D phonon transport simulation with SAAF method versus experimental results.

Self-Adjoint Formulation of the Phonon BTE

Applying the relaxation time approximation [6] to Eq. (2) allows for a manageable numerical solution of the steady state BTE for phonons. For brevity we have suppressed the independent variables in many of the terms of the equations: $f_\omega = f_\omega(\omega, p, \theta, \phi)$ is the phase space phonon frequency distribution, $\tau = \tau(\omega, p, \theta, \phi)$, $v_g = v_g(\omega, k)$. Here, ω is the phonon frequency, p is the phonon polarization (which is transverse or longitudinal), k is the wavevector, θ and ϕ are the polar and azimuthal angular components in spherical coordinates. The mean free path for phonons is defined as $\Lambda = v_g \tau$, the product of group velocity v_g and the phonon mean collision time between scattering events τ (relaxation time). To derive the three dimensional BTE for phonons in the self-adjoint form, some manipulation is required. Solving Eq. (2) for the phonon frequency distribution, and inserting the result back into Eq. (3) yields Eq. (4)

$$v_g \boldsymbol{\Omega} \cdot \boldsymbol{\nabla} f_\omega = \frac{f_\omega^0 - f_\omega}{\tau}, \qquad (2)$$

$$f_\omega = f_\omega^0 - \Lambda \boldsymbol{\Omega} \cdot \boldsymbol{\nabla} f_\omega. \qquad (3)$$

Expanding the remaining terms and isolating the two phonon distributions gives

$$\Lambda \boldsymbol{\Omega} \cdot \boldsymbol{\nabla} \left(f_\omega^0 - \Lambda \boldsymbol{\Omega} \cdot \boldsymbol{\nabla} f_\omega \right) = f_\omega^0 - f_\omega, \qquad (4)$$

$$-\Lambda \boldsymbol{\Omega} \cdot \boldsymbol{\nabla} \left[\Lambda \boldsymbol{\Omega} \cdot \boldsymbol{\nabla} f_\omega \right] + f_\omega = -\Lambda \boldsymbol{\Omega} \cdot \boldsymbol{\nabla} f_\omega^0 + f_\omega^0. \qquad (5)$$

Equation (5) is the BTE for phonons in the SAAF derivation. Here f_ω is the phonon frequency distribution, f_ω^0 is the equilibrium phonon distribution; both are governed by Bose-Einstein statistical mechanics. Equation (5) bears close resemblance to the SAAF for neutron transport, and with some small amount of work the Rattlesnake neutron transport code can be used to model phonon transport. Phonons and neutrons may both be classified as neutral particles, though phonons are created through temperature excitations in material; neutrons are ultimately conserved. Neutrons may fundamentally alter matter through absorption or fission, phonons may not. Neutrons have an inherent potential to be absorbed or scattered when interacting with a nucleus, contributing to a total cross section (probability of interaction); phonons interact primarily through scattering, based on their mean free path (Λ) in the material.

Rattlesnake is appropriate for calculations in purely scattering media. In addition, there is no external source as may be present in a neutron transport problem. Phonons are created entirely as a function of temperature and material physics. In this paper, we model a problem with temperature driven sources at the boundaries. The boundary phonon intensity is found through a relation of the group velocity, volumetric specific heat, and zone temperature: $I_b = \frac{C_v v_g T_b}{4\pi}$. When temperatures are much lower than the Debye temperature, boundary scattering of phonons dominates. This explains the decreased phonon distributions at the boundaries as seen in Fig. 1. The Debye temperature is associated with the Debye frequency, such that it is a cutoff frequency for different phonon density of states. The Debye temperature for silicon is approximately 645 K. We use temperature boundaries of $T_L = 301$ K and $T_R = 300$ K consistent with the published test problem [1, 2].

Conversion to Frequency Independence

We use the relaxation time approximation phonon BTE, but because we are interested in diffusive scattering behavior of phonons we must convert the result to the EPRT. The frequency independent phonon intensity $I(r, \theta, \phi)$ is defined as [1]

$$I(r, \theta, \phi) = \frac{1}{4\pi} \int_0^{\omega_{max}} \sum_p v_g f_\omega \hbar \omega(k) D(\omega) d\omega, \qquad (6)$$

where v_g is the phonon group velocity, f_ω is the phonon distribution function, $\hbar$ is the reduced Planck's constant, ω is the phonon frequency, $D(\omega)$ is phonon density of states and ω_{max} is an arbitrary limiting frequency. The SAAF BTE now becomes a transport equation for a gray medium. We achieve this by taking a summation over the polarization with respect to the group velocity, phonon density of states and multiplying by the frequency distribution.

The following equations will help clarify the source iteration process for the phonon BTE. As there is no traditional source term to initialize the calculation, we rely instead on the difference between phonon intensity and phonon equilibrium intensity, providing an initial guess and then subsequent refinement of that value. We do not iterate on a source, but instead iterate on the phonon equilibrium intensity. Assuming steady state and no internal heat generation, we set $\boldsymbol{\nabla} \cdot q = 0$ and remove this term from the governing equation. Thus,

$$\int_0^{2\pi} \int_0^\pi I^0(r, \theta, \phi) \sin\theta d\theta d\phi$$

$$= \int_0^{2\pi} \int_0^\pi I(r, \theta, \phi) \sin\theta d\theta d\phi, \qquad (7)$$

where I is phonon intensity, and I^0 is the equilibrium phonon intensity. With this relationship, it is possible to determine temperatures at both locations. As the equilibrium intensity is only a function of space, it may be taken outside the integral. Thus Eq. (8) provides the source iteration component of the discretization scheme

$$I^0(r) = \frac{1}{4\pi} \int_0^{2\pi} \int_0^\pi I(r, \theta, \phi) d\Omega. \qquad (8)$$

The extracted temperature from the equilibrium intensity represents an average energy of total phonons at a local point. This is comparable to the equilibrium temperature of phonons as they undergo adiabatic repopulation to an equilibrium state. Temperature can be found from the phonon intensity by integrating the cell intensity over solid angle;

$$T(r) = \frac{1}{C_v v_g} \int_0^{2\pi} \int_0^\pi I(r, \theta, \phi) d\Omega. \qquad (9)$$

TEST PROBLEM DESCRIPTION

The test problem selected was one from the experiment of Yilbas and Bin Mansoor [2]. They investigated the effect of phonon transport in a thin film of silicon. They derived

the BTE for phonons using a finite difference scheme. Data for this test problem was taken from Majumdar [1], and is presented in Table I.

The problem from [2] was simulated in Rattlesnake, with the geometry constructed using a Python script in Cubit. A cube of silicon with side lengths of $L = 101.4$ nm (3Λ) was generated and a square mesh was applied; both fine and coarse meshes were used. Reflecting conditions were imposed on planes in XY and XZ domains, and planes in YZ were given a temperature boundary source condition. This step simulates one dimensional transport. We simulate a steady-state problem with isotropic scattering and assume that all phonons have the same group velocity. The solver type is a Preconditioned Jacobian Free Newton Krylov (PJFNK) using *boomeramg* and *hypre* options in PETSc [7]. An S_{16} angular quadrature set is used. The problem executed with 2 nonlinear iterations, and a run time of 335 s using the fine mesh.

TABLE I. Physical Data

$C\ (\mathrm{J \cdot m^{-3} \cdot K^{-1}})$	Λ (m)	$v_g\ (\mathrm{m \cdot s^{-1}})$	$T_L\ (K)$	$T_R\ (K)$
$1.653 \cdot 10^6$	$33.8 \cdot 10^{-9}$	8430	301	300

RESULTS

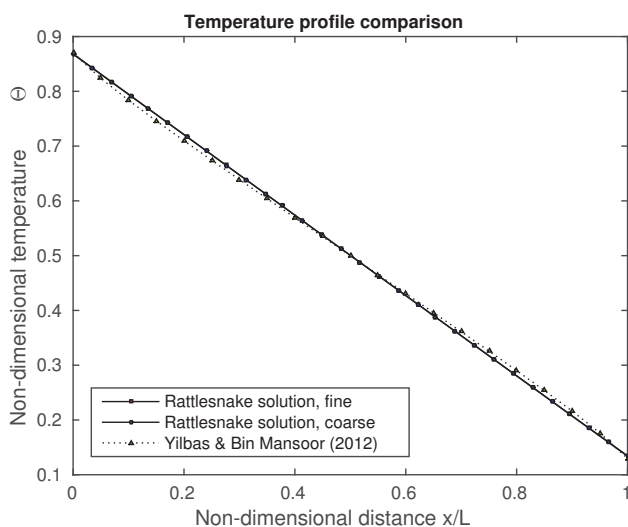**Test Problem - Homogeneous Medium**

Fig. 2. Rattlesnake output versus experiment in [2]

Fig. 2 compares transport results in Rattlesnake to that of the published data. It is evident from the figure and analysis that the output is in very good agreement with Yilbas and Bin Mansoor. The results of the coarse and fine mesh are nearly identical, but the coarse mesh problem had a runtime an order of magnitude smaller than that of the fine mesh.

Because of the steady state assumption, there is zero transient heat flux; this is evident from Fig. 2. The shape of the curve from [2] is slightly nonlinear, with a depression above the midpoint and a subsequent inflection below. The plot of the Rattlesnake solution appears to be sharply linear in contrast. This difference is likely due to the higher fidelity of the Rattlesnake calculation, with the increased availability of quadrature angles. The finite element discretization scheme as opposed to the finite difference scheme used in [2] may also have an effect.

An important feature to note in Fig. 2 are the intercepts at both zero and unity of the non-dimensional distance. Intuition suggests that they should intercept at 1 at the left hand side and 0 at the right hand side due to the imposed temperature boundaries. However, because the thickness of the cube is three mean free paths boundary scattering of phonons dominates when thicknesses shrink to the order of the mean free path of phonons in the material; materials of this kind are said to approach the Casimir limit [8].
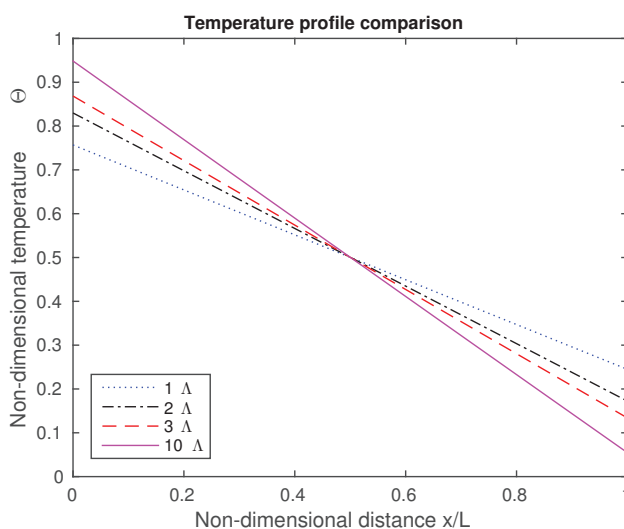

Fig. 3. Acoustic thickness comparison

At the boundaries of Fig. 3, further evidence is presented to show discontinuity between non-dimensional temperature values and prescribed temperature boundary conditions. The expected solutions that would correspond to $T_L = 301$ K and $T_R = 300$ K are actually shifted on each border due to boundary scattering effects. This is apparent from the lines of Fig. 3, which are four silicon cube geometries with varying acoustic thickness. It is evident from these variations that as the geometry enlarges or shrinks to approach the Casimir limit, the phonon intensity shifts and results in step changes at the boundaries. As the problem models a purely scattering medium, some of the phonon flux is reflected outright rather than contribute to the magnitude at the opposing plane. The closer the emissive planes, the more pronounced the effect. This phenomenon is responsible for the shifted boundary values in Fig. 3.

2D Heterogeneous Problem

We have simulated a two-dimensional plane of silicon with a small square of aluminum in the center in order to show the heterogeneous medium phonon transport capabilities of Rattlesnake. The dimensions of the square are 250 nm per

side. The aluminum square sits at the center of the silicon, and is 3.4 nm per side. The boundaries of the aluminum and silicon were set at 301 K and 300 K, respectively. All temperature boundaries provide effective phonon intensities and were simulated at steady state. Physical data for aluminum was obtained from [9].
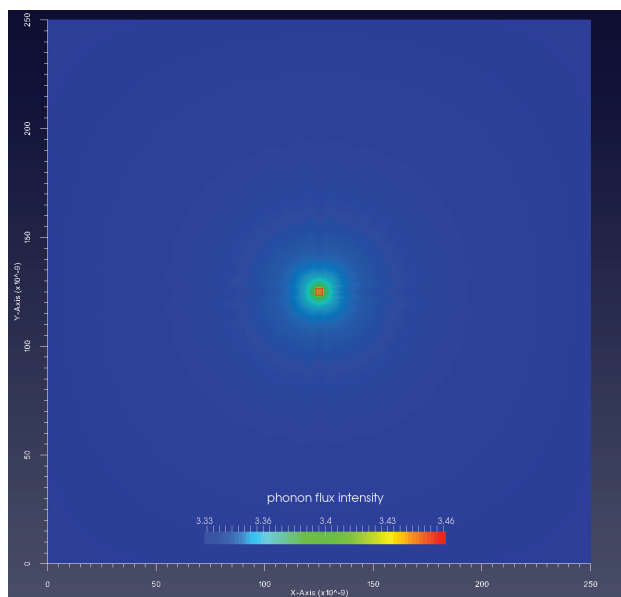

Fig. 4. Silicon square with an aluminum impurity

Fig. 4 shows the effect of the heated aluminum square on surrounding silicon. The temperature of the aluminum is assumed to be spatially uniform. A temperature jump exists at the boundary of aluminum and silicon, which is the response due to thermal boundary resistance at this interface. Diffusive transport behavior is seen as heat dissipates into the surrounding body of silicon, which was set at 300 K and acts as a heat sink. As phonons are transported from the aluminum heat source into the surrounding bulk, quasi-ballistic behavior also begins to have an effect. Further out towards the boundaries of Fig. 4, ray effects are observed, which are an artifact of the use of the discrete ordinates angular discretization.

CONCLUSION

We have shown that the BTE for phonons may be derived in the SAAF formulation, and that transport solutions using Rattlesnake are consistent with those of previous studies which used different methods [1, 2]. Our solutions also compare well with the exact solution of the EPRT.

Rattlesnake is able to capture different regimes of phonon scattering, exhibiting agreement with published data for diffusive and ballistic behaviors. Mesh and angular quadrature studies may be performed to optimize the trade-off between accuracy and speed of solution. More complicated problems and geometries will be constructed to explore the full capability of the transport engine. Diffusion synthetic acceleration (DSA) might be employed to further expedite the solution. Rattlesnake has a nonlinear diffusion acceleration (NDA) capability and we plan to investigate this in the future.

Phonon transport with Rattlesnake could lead to development of a tool within the MOOSE family to calculate thermal conductivity in nuclear fuel, when provided with initial temperatures and material data. This tool may be used in conjunction with other simulation packages which provide parameters such as relaxation times and phonon group velocities to compute the mean free path. Knowledge of thermal conductivity in nuclear fuels is imperative for the continued safe operation of nuclear power plants and research reactors, and may prove beneficial in the further development and deployment of Generation IV nuclear reactors.

REFERENCES

1. A. MAJUMDAR, "Microscale Heat Conduction in Dielectric Thin Films," *Journal of Heat Transfer*, **115**, *7*, 7–16 (1993).
2. B. YILBAS and Y. BIN MANSOOR, "Phonon Transport in Two-Dimensional Silicon Thin Film: Influence of Film Width and Boundary Conditions on Temperature Distribution," *European Physical Journal B*, **85**, 234–242 (2012).
3. E. A. GASTON, D., "MOOSE: A Parallel Computational Framework for Coupled Systems of Nonlinear Equation," *Nuclear* (2009).
4. J. MOREL and J. MCGHEE, "A Self-Adjoint Angular Flux Equation," *Nuclear Sci*, **3**, *12*, 312–325 (1999).
5. *MATLAB and Statistics Toolbox Release 2014b, The Math-Works, Inc., Natick, Massachusetts, United States.*
6. C. KITTEL, *Introduction to Solid State Physics*, John Wiley & Sons, Inc., 8th ed. (2005).
7. S. BALAY, S. ABHYANKAR, M. F. ADAMS, J. BROWN, P. BRUNE, K. BUSCHELMAN, V. EIJKHOUT, W. D. GROPP, D. KAUSHIK, M. G. KNEPLEY, L. C. MCINNES, K. RUPP, B. F. SMITH, and H. ZHANG, "PETSc Users Manual," Tech. Rep. ANL-95/11 - Revision 3.5, Argonne National Laboratory (2014).
8. H. B. G. CASIMIR, "Notes on the Conduction of Heat in Crystals," *Physica*, **5**, *6*, 495–500 (1938).
9. B. YILBAS and Y. BIN MANSOOR, "Lattice Phonon and Electron Temperatures in Silicon-Aluminum Thin Films Pair: Comparison of Boltzmann Equation and Modified Two-Equation Model," *Journal of Computational and Theoretical Transport* (2014).