Effective Thermal Conductivity of Tri-Isotropic (TRISO) Fuel Compacts

Charles P. Folsom
Utah State University

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EFFECTIVE THERMAL CONDUCTIVITY OF
TRI-ISOTROPIC (TRISO) FUEL COMPACTS

by

Charles Folsom

A thesis submitted in partial fulfillment
of the requirements for the degree

of

MASTER OF SCIENCE

in

Mechanical Engineering

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2012
ABSTRACT

Effective Thermal Conductivity of Tri-Isotropic (TRISO) Fuel Compacts

by

Charles P. Folsom, Master of Science
Utah State University, 2012

Major Professor: Dr. Heng Ban
Department: Mechanical and Aerospace Engineering

Thermal conductivity is an important thermophysical property needed for effectively predicting nuclear fuel performance. As part of the Next Generation Nuclear Plant (NGNP) program, the thermal conductivity of tri-isotropic (TRISO) fuel needs to be measured over a temperature range characteristic of its usage. The composite nature of TRISO fuel requires that measurement be performed over the entire length of the compact in a non-destructive manner. No existing measurement system is capable of performing such a measurement.

A measurement system has been designed based on the steady-state, guarded-comparative-longitudinal heat flow technique. The system is capable of measuring cylindrical samples with diameters ~12.3 mm (~0.5 in.) with lengths ~25 mm (~1 in.). The system is currently operable in a temperature range of 100–700°C for materials with thermal conductivities on the order of 10–70 W·m⁻¹·K⁻¹. The system has been designed, built, and tested. An uncertainty analysis for the determinate errors of the system has been performed finding a result of 6%.
Measurements have been performed on three calibration/validation materials: a certified glass ceramic reference material, 99.95% pure iron, and Inconel 625. The deviation of the validation samples is < 6-8% from the literature values. In addition, surrogate NGNP compacts and NGNP graphite matrix-only compacts have been measured. The results give an estimation of the thermal conductivity values that can be expected. All the results are presented and discussed.

A Finite Element Analysis was done to compare the accuracy of multiple effective conductivity models. The study investigated the effects of packing structure, packing fraction, matrix thermal conductivity, and particle heat generation. The results show that the Maxwell and the Chiew & Glandt models provide the most accurate prediction of the effective thermal conductivity of the TRISO fuel compacts.

Finally, a discussion of ongoing work is included as well as the possibility of correlating effective thermal properties of fuel compacts to their constituents with measurements of well-defined samples.
PUBLIC ABSTRACT

Effective Thermal Conductivity of Tri-Isotropic (TRISO) Fuel Compacts

Charles Folsom

The Next Generation Nuclear Plant (NGNP) program objective is to develop a new type of nuclear reactor that produces process heat instead of electricity. The process heat can be used in the production of hydrogen and many other industrial processes. As part of the NGNP program a new type of nuclear fuel is being developed. The fuel is a composite construction of specially coated fuel particles and graphite pressed together in a cylindrical compact.

Thermal conductivity is an important thermophysical property of the fuel that needs to be measured. Knowledge of the thermal conductivity of the fuel will provide accurate prediction of fuel performance and safety assessment of the nuclear reactor. The composite nature of the fuel compact requires the thermal conductivity measurement be performed over the entire length of the compact in a non-destructive manner. No existing measurement system is capable of performing such a measurement.

The objective of this study was to characterize the thermal conductivity of the fuel compact. This was done both experimentally and numerically. Previously a measurement system was built to measure the thermal conductivity of the fuel compact. That measurement system had to be validated to ensure the accuracy of the measurements. Once the system was validated it was used to measure multiple surrogate fuel compacts. The numerical work was performed using a finite element model developed in COMSOL Multiphysics. The purpose of the numerical model...
was to compare the thermal conductivity of the fuel compacts to previously established models that predict the thermal conductivity of two phase composites.

The results from the experimental study provide an estimate of the expected thermal conductivity of the fuel compacts. The numerical results provided a possible model that can be used to predict the thermal conductivity of the nuclear fuel compacts.
ACKNOWLEDGMENTS

I would like to express my sincere gratitude to my major professor, Dr. Heng Ban, who provided me the opportunity to pursue my postgraduate degree at Utah State University and who continuously provided the encouragement and support needed to accomplish this work. The guidance he has provided me in my research and life has been a great influence to me. I would also like to thank Dr. Changhu Xing and Colby Jensen for their help, encouragement, and friendship throughout this work. I would also like to thank all the members of the Multiscale Thermophysics Lab at Utah State University for their help and especially friendship. Lastly I will be eternally grateful to my parents, John and Tracy Folsom, for their love and encouragement throughout my life.

Charles Folsom
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<td>American Society for Testing and Materials</td>
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<tr>
<td>B&amp;W</td>
<td>Babcock &amp; Wilcox</td>
</tr>
<tr>
<td>BCC</td>
<td>body centered cubic</td>
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<tr>
<td>EMT</td>
<td>Effective Medium Theory</td>
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<tr>
<td>ETC</td>
<td>Effective thermal conductivity</td>
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<td>FCC</td>
<td>face centered cubic</td>
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<td>FEA</td>
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<td>HTGR</td>
<td>High Temperature Gas Reactor</td>
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<td>IRMM</td>
<td>Institute for Reference Materials and Measurements</td>
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<td>NGNP</td>
<td>Next Generation Nuclear Plant</td>
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<td>ORNL</td>
<td>Oak Ridge National Laboratory</td>
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<td>PARFUME</td>
<td>PArticle Fuel ModEl</td>
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<tr>
<td>SC</td>
<td>simple cubic</td>
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<td>TRISO</td>
<td>tri-structural isotropic</td>
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<td>TSM</td>
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\( k_{so} \) Corrected thermal conductivity of test specimen, \([W \cdot m^{-1} \cdot K^{-1}]\)

\( k_{100} \) Fitting parameter for German matrix thermal conductivity

\( L \) Height of unit cell, [m]

\( N \) A parameter for the Zehner and Schlünder model

\( q \) Heat flow, [W]

\( q_m \) Heat flow in meter bar, [W]

\( q_s \) Heat flow in test sample, [W]

\( \dot{q} \) Volumetric heat generation, \([W/m^3]\)

\( q'' \) Heat flux, \([W/m^2]\)

\( q_{z''} \) Heat flux in the z direction, \([W/m^2]\)

\( r \) Particle radius, [mm]

\( s \) Side of rectangular prism used for the random model, [mm]

\( s_i \) Random standard uncertainty for a parameter “i”

\( T \) Generic temperature, [K]

\( T' \) Fitting parameter for German matrix thermal conductivity

\( T_b \) Temperate at bottom of unit cell, [K]

\( T_t \) Temperature at top of unit cell, [K]

\( U \) Uncertainty

\( x \) Cartesian coordinate system direction

\( y \) Cartesian coordinate system direction

\( z \) Cartesian coordinate system direction

\( \dot{q} \) Volumetric heat generation, \([W/m^3]\)

\( T_1 \) Average temperature of first meter bar, [K]

\( \bar{T}_2 \) Average temperature of second meter bar, [K]
Average temperature of test sample, [K]

Fitting parameter for German matrix thermal conductivity

Reduced thermal polarizability

Neutron fluence

Fitting parameter for German matrix thermal conductivity

A parameter for the Lewis and Nielsen model

Generic temperature difference, [K]

Temperature difference between two temperature monitors on the meter bars, [K]

Temperature difference from two temperature monitors on the test specimen, [K]

Measured temperature difference across first meter bar, [K]

Measured temperature difference across second meter bar, [K]

Generic position difference, [m]

Distance between temperature monitor positions in first meter bar, [m]

Distance between temperature monitor positions in second meter bar, [m]

Distance between two temperature monitor positions on the meter bars, [m]

Distance between temperature measurement points in test sample, [m]

Ratio of particle thermal conductivity to matrix thermal conductivity

A parameter for the Lewis and Nielsen model

Particle volume fraction

Maximum packing fraction of dispersed particles

Density, [kg/m$^3$]

A parameter for the Lewis and Nielsen model
CHAPTER 1

INTRODUCTION

There is a need, as part of the Next Generation Nuclear Plant (NGNP) program, to characterize the material properties of the fuel. One of these properties, thermal conductivity, is necessary in order to correctly model the fuel performance. The fuel consists of coated fuel particles of approximately 1 mm diameter sintered together in a graphite matrix. The final fuel “compact” is cylindrically shaped and approximately 12.3 mm (0.5 inch) in diameter by 25 mm (1 inch) in length [1]. The coated particles consist of a fuel kernel that is coated with tri-structural isotropic (TRISO) layers: a porous pyrolytic carbon layer (buffer layer), pyrolytic carbon (inner) layer, silicon carbide layer, and pyrolytic carbon (outer) layer [2] (see Figure 1-1 for a representation of the TRISO particle). Since the fuel compacts are a non-homogenous mixture of multiple materials, the thermal conductivity required is an effective thermal conductivity (ETC) of all the materials in the fuel compact.

Figure 1-1. TRISO particles with labeled layers
An understanding of the thermophysical properties of the fuel is crucial for predictive capability and modeling. Understanding the properties of the fuel will also ensure that the fuel is operating at appropriate temperatures to ensure safe operation of the reactor. To date, the effective thermal conductivity of the TRISO fuel compact has not been measured and therefore and accurate value for the ETC is not currently known. The ability to model the fuel will provide valuable information on the fuel conditions and performance that exist in the reactor. A change in the type or amount of any of the materials will have an effect upon the effective conductivity of the fuel compact, and thus the conditions that exist in the reactor.

Currently the modeling of the fuel uses a code called PARFUME (PARticle Fuel ModEl). PARFUME is being used as an advanced gas-cooled reactor fuel performance and analysis code [3]. The thermal conductivity values used by the code are from matrix-only data measured by the Germans in the 1980’s [4]. To account for the effect of the particles in the fuel compact, a correction factor [3] is applied to the matrix-only thermal conductivity value. This correction factor ($F_p$) developed by the Germans can be seen in Equation (1.1), where $\phi$ is the particle packing fraction by volume. This correction factor developed by the Germans is just a reduced form of the effective conductivity model derived by Maxwell [5] in 1881. Equation (1.2) is the full version of the Maxwell’s equation where $k_m$ is the matrix conductivity and $k_p$ is the particle conductivity. If the particle thermal conductivity is zero then Equation (1.2) reduces to Equation (1.1). So the reduced form of Maxwell’s equation treats the particles as voids in the matrix material. There are a lot of other effective conductivity models that have been developed over the years and it is possible that one of these other models represents the ETC of the fuel better than Equation (1.1). The models that are more appropriate for the fuel compacts will be discussed later, but a good review of effective conductivity models was performed by Progelhof [6].

As part of this report a Finite Element Analysis (FEA) study was performed to compare the results between the full Maxwell’s equation, the reduced form, and other popular effective
conductivity models. It will be shown later in the report that larger errors can occur when using Equation (1.1). The large difference between the thermal conductivity of the fuel and the estimated thermal conductivity from the German data and Equation (1.1) can result in large discrepancies in the reactor conditions. If the PARFUME model over predicts the thermal conductivity of the fuel then the temperatures in the reactor will be higher than what the PARFUME model determines. This could result in unsafe conditions. Therefore it is important for safety and also efficiency concerns that the correct thermal conductivity values are used in the modeling of the NGNP reactor.

\[
F_p = \frac{1 - \phi}{1 + \frac{\phi}{2}} \quad (1.1)
\]

\[
k_e = \frac{3k_m k_p \phi + (2k_m + k_p) k_m (1 - \phi)}{3k_m \phi + (2k_m + k_p)(1 - \phi)} \quad (1.2)
\]
2.1. Thermal Conductivity Measurement System

2.1.1. Background

There are many thermal conductivity measurement methods that exist [7–11]. Each method has advantages and disadvantages that can limit its use for certain applications. The main factors when selecting a thermal conductivity measurement method include [12]:

- Expected thermal conductivity of the sample
- Size and geometry of the sample
- Required temperature range
- Magnitude of temperature gradient
- Accuracy required
- Electrical conductivity of the sample
- Fabrication difficulties
- Measurement time
- Density and specific heat of the sample
- Level of porosity
- Inhomogeneities in the material (e.g., composite materials)

Because of the cylindrical shape, the expected medium-to-high thermal conductivity of the fuel compacts, and the need for a non-destructive measurement method, an axial heat flow method was selected. Also due to the small size and the desired temperature range of measurements of the sample, the comparative axial heat flow technique [13] was selected.
The comparative axial heat flow method is a comparative, steady-state method. It has been around since the 1930s [14,15] and was studied extensively in the 1950s and 60s by Ballard [15], Morris and Hust [16], Franel and Kingery [17], and Mirkovich [18]. The claimed accuracy of the system was questioned by Laubitz [19], but later studies by Sweet et al. [20] and Pillai and George [21] reported accuracies, independent of the uncertainty of the reference sample, to be better than ±5%. The American Society for Testing and Materials (ASTM) produced a standard for this method in 1987, ASTM E 1225 [22], which was revised in 2004. Also Babelot [23] performed tests with a commercial comparative thermal conductivity apparatus that was used in a glovebox.

2.1.2. Method

The experimental measurement system was designed for the TRISO fuel geometry, expected thermal conductivity values (10-70 W·m⁻¹·K⁻¹), and expected operating temperature range (100-700°C) of the fuel compacts. Much of the design is based upon details from Didion [24] and the ASTM E 1225 Guarded Comparative Longitudinal Heat Flow Technique [22]. Figure 2-1 shows a schematic of the experimental measurement system and the main components used, and Figure 2-2 shows a photo of the actual measurement system. A schematic of the actual measurement section of the measurement system can be seen in Figure 2-3. Figure 2-4 shows a photograph of the assembled measurement section and Figure 2-5 shows the measurement section inside the tube furnace. The primary functions of the measurement section include:

1. Create a controlled, steady-state temperature gradient through the sample and meter bars. By surrounding the meter bars and test sample with insulation as well as a temperature controlled guard, the heat flow can be directed through the sample column (meter bars and test sample).
2. Create reproducible conditions in the measurement section. This is accomplished with the use of a spring to apply a constant force and thus a constant contact pressure at the interfaces of the sample and meter bars.

3. Measure steady-state temperature gradients in the sample and meter bars.

Figure 2-1. Schematic overview of the experimental measurement system.

Figure 2-2. Measurement system.
Figure 2-3. Schematic of the measurement section of the measurement system.

Figure 2-4. Assembled measurement section.

Figure 2-5. Measurement section in tube furnace.

Note: for a more detailed explanation of the measurement system design please refer to the work performed by Jensen [25, 26].
2.1.3. Theory

The technique used to measure the ETC of the fuel compacts is the guarded-comparative-longitudinal heat flow method. With this technique a sample of unknown thermal conductivity is sandwiched between two reference samples of known thermal conductivity (Figure 2-6). The steady-state temperature gradients are measured with thermocouples placed at precise positions. The temperature gradients are used to calculate the thermal conductivity.

![Schematic of the guarded-comparative-longitudinal heat flow technique](Image)

Figure 2-6. Schematic of the guarded-comparative-longitudinal heat flow technique.

The thermal conductivity is calculated using Fourier’s Law,

\[ q''_l = -k \frac{dT}{dz}, \]  

(2.1)
where $q_z$ is the heat flux in the axial direction, $k$ is the thermal conductivity, and $dT/dz$ is the temperature gradient. From the measured temperature gradients ($\Delta T/\Delta Z$), cross-sectional area ($A_m$), and thermal conductivity at the average temperature of the meter bars ($k_{mb}$), the heat flow through each meter bar can be calculated. The heat flow through the sample ($q_s$) is calculated as the average of the heat flows in the meter bars. The equation used to calculate the average heat flow is,

$$q_s = \frac{k_{mb}(T_1)A_m \frac{\Delta T_1}{\Delta Z_m} + k_{mb}(T_2)A_m \frac{\Delta T_2}{\Delta Z_m}}{2}. \quad (2.2)$$

Using the calculated heat flow in the sample from Equation (2.2) and the measured temperature gradient of the sample, the ETC of the sample can be calculated as

$$k_s(T_s) = q_sA_s \frac{\Delta T_s}{\Delta T_s} \quad (2.3)$$

The calculated thermal conductivity of the sample from Equation (2.3) is the thermal conductivity at the average temperature of the sample.

2.1.4. Thermal Conductivity Correction

The ASTM E 1225 standard [22] for this measurement method suggests that corrections be applied to the thermal conductivity to account for extraneous heat flow. The correction is especially important when the specimen, meter bars, or both have a low thermal conductivity relative to the insulation. The standard suggests three possible ways to apply corrections to the calculated thermal conductivity:

1. Use analytical techniques described by Didion [24] and Flynn [27].
2. Using calculations from finite-difference or finite-element heat conduction codes.
3. Determined experimentally by using reference materials that are the same size as the test specimen and have the same surface finish.
The correction procedure used will be discussed in detail later as this was one of the main objectives of this work.

2.2. Effective Thermal Conductivity Models

2.2.1. Background

A major interest in this work is to be able to find a theoretical model from which the ETC of the composite fuel compacts may be determined from their constituents and compositions. There are a large number of models in literature that have been derived for predicting the ETC of heterogeneous materials. Reviews of such models can be found in multiple sources [6, 28–33]. Even with the great number of models available, the uncertainty in predicting the ETC of heterogeneous materials can still be quite high [34]. Many of the models are for specific materials and/or compositions. Some require empirically derived parameters. For these reasons, the data collected from the thermal conductivity measurement system will be used to validate a model for the NGNP compacts and as a direct comparison between various materials and material conditions.

The parameters affecting the ETC in heterogeneous materials are well defined by Tsotsas [28]. In the case of the NGNP fuel compacts where the graphite matrix is considered continuous and the particles are uniform in size, the ETC, $k_e$, is a function of: the thermal conductivity of the graphite matrix, $k_m(T, \Gamma)$, the thermal conductivity of the particles (the TRISO particles are themselves a composite, so the ETC will need to be determined for the particles as well), $k_p(T, \Gamma)$ (which are also functions of temperature, $T$, and neutron fluence, $\Gamma$), the particle volume fraction, $\phi$, and the particle arrangement, $a$.

$$k_e = k_e(k_m(T, \Gamma), k_p(T, \Gamma), \phi, a)$$  \hspace{1cm} (2.4)
For this work the effect on the thermal conductivity due to the neutron fluence will not be investigated, but the parameters of interest will be the thermal conductivity of the matrix and particle, the packing fraction, and the packing arrangement.

2.2.2. Historical Work

In the 1980’s, Gontard and Nabeilek [4] presented an empirical model developed for calculating the ETC of German TRISO fuels. The model defines the thermal conductivity of the matrix by

\[ k(T) = k_{100}(1 - \alpha T' e^{\delta T}), \]  

where \( T' = T - 100^\circ C \), \( \alpha \) and \( \delta \) are constants, and \( k_{100} \) is the thermal conductivity at \( 100^\circ C \). For the A3-27 material used for the PARFUME modeling the constants are:

\[ k_{100} = 47.4 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}, \]
\[ \alpha = 9.7556 \cdot 10^{-4}, \]
\[ \delta = -6.0360 \cdot 10^{-4}. \]

The model included a factor (Equation (1.1)) to account for the particle volume fraction, as well as neutron fluence. The factor for particle volume fraction is simply a simplified version of Maxwell’s equation for effective conductivity, which will be discussed later. Figure 2-7 shows a plot of the thermal conductivity of the A3-27 German matrix material as well as the ETC for 0.30, 0.35, and 0.40 particle volume fractions using the factor (Equation (1.1)).
2.2.3. Analytical Models

In the interest of finding a correlating model for the ETC of NGNP fuel, a brief summary of relevant work will be presented. Some of the fundamental models are presented first followed by some models that are well established. Two useful parameters used in ETC studies are the ratio of the thermal conductivity of the dispersed phase (particles) to the continuous matrix,

$$\kappa = \frac{k_p}{k_m},$$  \hspace{1cm} (2.6)

and the reduced polarizability,

$$\beta = \frac{\kappa - 1}{\kappa + 2},$$ \hspace{1cm} (2.7)

Figure 2-7. German TRISO fuel thermal conductivity showing the effect of particle volume fraction. Matrix material is A3-27 graphite heat-treated to 1800°C.
2.2.3.1. Series and Parallel

The minimum and maximum effective thermal conductivities for a two phase composite are given by the series and parallel phase distributions [35]. The minimum value occurs with the series distribution where the two phases are in layers normal to the direction of heat flow. The ETC for the series distribution is given by:

\[ k_e = (1 - \phi)k_m + \phi k_p. \]  

The maximum value occurs when the two phases are in layers parallel to the direction of heat flow. The ETC for the parallel distribution is given by:

\[ k_e = \frac{k_m}{\phi \left( \frac{1}{k} - 1 \right) + 1}. \]  

2.2.3.2. Geometric Mean

The geometric mean model [36] assumes a random distribution of the two phases. The ETC is calculated based upon a weighted geometric mean of the conductivities of the two constituents. The model is given by:

\[ k_e = k_p^\phi k_m^{1 - \phi}. \]  

2.2.3.3. Maxwell’s Equation

Maxwell’s equation is one of the oldest and probably the most well-known effective conductivity model. Many of the models since Maxwell’s have been based upon this model [37]. Using potential theory he derived an exact solution for the conductivity of homogeneous spheres in a homogeneous continuous medium [6]. Maxwell’s model makes no assumptions to the geometric configuration of the particles [38], but it does assume that the particles are far enough apart so that the temperature distortions due to the particles do not interfere with the surrounding
particle temperature distributions [6, 37, 39]. Maxwell’s equation can be seen in Equation (1.2), or using Equations (2.6) and (2.7) it can be written as:

\[
\frac{k_e}{k_m} = \frac{1 + 2\beta \phi}{1 - \beta \phi}.
\] (2.11)

Phase Inverted Maxwell’s Equation

The regular Maxwell’s equation is used when the thermal conductivity of the dispersed phase is less than the conductivity of the continuous phase (\(\kappa < 1\)). When the conductivity of the dispersed phase is larger than the continuous phase (\(\kappa > 1\)), then the phase inverted Maxwell’s equation is appropriate [5]:

\[
\frac{k_e}{k_m} = \frac{(1 + 2\beta \phi)(1 - \beta + 2\beta \phi)}{(1 - \beta)(1 + 2\beta - \beta \phi)}.
\] (2.12)

Reduced Maxwell’s Equation (German Correction)

The reduced form of Maxwell’s equation is not very common, but it is included for comparison because it is the effective conductivity model used for the PARFUME modeling. The reduced form of Maxwell’s equation is derived by first starting with Equation (1.2), then by setting \(k_p = 0\) it reduces to

\[
\frac{k_e}{k_m} = \frac{2(1 - \phi)}{2 + \phi}.
\] (2.13)

Equation (2.13) becomes Equation (1.1) by setting \(F_p = k_e/k_m\). 

2.2.3.4. Hashin and Shtrikman

Hashin and Shtrikman [40] proposed the two most restrictive bounds for ETC of a two phase material where the dispersed phase has a spherical geometry. The bounds actually correspond to Maxwell’s equations for a dilute dispersion (\(\phi \ll 1\)) [41]. For \(\kappa < 1\), the regular and phase inverted Maxwell equations describe the upper and lower bounds respectively. For
\( \kappa > 1 \), the phase inverted and regular Maxwell equations describe the upper and lower bounds, respectively.

2.2.3.5. Effective Medium Theory

The Effective Medium Theory (EMT) model is also a very popular model. Just as many of the effective conductivity models have been based upon Maxwell’s equation, a lot have also been based on the EMT model [37]. The EMT model makes no assumption about the shape of the particles and also assumes a random distribution of the particles [34, 37, 39]. The most common version of the EMT model is the Bruggeman model which treats the local conductivities as fluctuations about the conductivity of a uniform medium [42]. In this model there is no distinction between the continuous (matrix) and dispersed (particles) phases, and is generally considered being more valid for \( \phi \gg 0 \). The Bruggeman form, which has shown to be in good agreement with data for several solid-solid compounds [43], of the EMT model is given as:

\[
\frac{k_e}{k_m} = \kappa A + \sqrt{\kappa^2 A^2 + \kappa/2},
\]  

(2.14)

and \( A \) is defined as:

\[
A = \frac{1}{4} (3\phi - 1 + (2 - 3\phi)\kappa^{-1}).
\]  

(2.15)

2.2.3.6. Zehner and Schlünder

The Zehner and Schlünder model is a commonly mentioned model [44]. It was derived assuming particle to particle contact using an analogy between mass transfer experiments and thermal conduction to obtain an empirical curve for \( 0 < \phi < 1 \). The ETC for Zehner and Schlünder is based on a unit cell and is given as:

\[
\frac{k_e}{k_m} = 1 + \sqrt{\phi(C - 1)},
\]  

(2.16)

where \( C \) is defined as
and N and B are:

\[ N = 1 - \frac{B}{\kappa} \]  \hspace{1cm} (2.18)

\[ B = 1.25 \left( \frac{\phi}{1 - \phi} \right)^{0.7}. \]  \hspace{1cm} (2.19)

Hsu [45] later found that more accurate results can be obtained by using:

\[ B = 1.364 \left( \frac{\phi}{1 - \phi} \right)^{1.055}. \]  \hspace{1cm} (2.20)

2.2.3.7. Lewis and Nielsen

In the review performed by Progelhof et al. [6], they showed that the Lewis and Nielsen [46] equation fitted the experimental data best for solid-solid composites. The shape and nature of packing of the dispersed particles is accounted for in their model. The Lewis and Nielsen model for spherical particles is:

\[ \frac{k_\varepsilon}{k_m} = \frac{1 - D \ E \ \phi}{1 - E \ \psi \ \phi'} \]  \hspace{1cm} (2.21)

where \( D = 1.5 \) for spheres and \( E \) and \( \psi \) are defined as:

\[ E = \frac{\kappa - 1}{\kappa + D'} \]  \hspace{1cm} (2.22)

\[ \psi = 1 + \frac{1 - \phi_m}{\phi_m^2} \phi. \]  \hspace{1cm} (2.23)

The term \( \phi_m \) is the maximum packing fraction of the dispersed particles. For hexagonal close and face-centered cubic packing fractions then \( \phi_m = 0.7405 \). Body-centered cubic and simple cubic packing fractions will result in \( \phi_m \) equal to 0.60 and 0.524, respectively. Lewis and Nielson also suggest that for a random close packing fraction then \( \phi_m = 0.637 \) and for a random loose packing fraction then \( \phi_m = 0.601 \).
2.2.3.8. Chiew and Glandt

The Chiew & Glandt model is an improved form of Maxwell’s equation that is correct to order $\phi^2$. This model matched experimental data for materials with $\kappa$ ranging from $10^{-3}$ to $10^4$ and $\phi$ from 0.15 to 0.85 very well. Using the best fitting parameters by Gonzo [47], the Chiew & Glandt model becomes:

$$\frac{k_e}{k_m} = \frac{1 + 2\beta\phi + (2\beta^3 - 0.1\beta)\phi^2 + \phi^3 0.05 \exp(4.5\beta)}{1 - \beta \phi} \quad (2.24)$$

2.2.3.9. Samantray

More recently, Samantray et al. [32] proposed a model to take into account the wide range of $\kappa$ and $\phi$. They predict the ETC based on the unit-cell approach (constant isotherms) as well as the semi-empirical field solution. Their model was compared to experimental values for ETC for a wide range of $\kappa$ and $\phi$ and proved to be more accurate than earlier proposed models. For values of $\kappa < 20$ and $0.1 < \phi < 0.9$, using the unit cell model developed by Raghaven and Martin [43], they show the ETC to be:

$$\frac{k_e}{k_m} = 1 + \frac{h}{1 + \zeta h} \quad (2.25)$$

where $h$ and $\zeta$ are:

$$h = -\frac{1}{\zeta} + \frac{1}{\zeta^2 \Delta \log \left(\frac{1 - \zeta (K_{\text{min}} - 1)}{1 - \zeta (K_{\text{max}} - 1)}\right)} \quad (2.26)$$

$$\zeta = \frac{1 - \beta - 3\beta\phi}{3\beta\phi} \quad (2.27)$$

And $\Delta = K_{\text{max}} - K_{\text{min}}$, where $K_{\text{min}}$ is Maxwell’s equation as defined in Equation (2.11), and $K_{\text{max}}$ is the phase inverted form of Maxwell’s equation as defined in Equation (2.12).
2.2.4. **Numerical Model**

In addition to the analytical ETC models another way to determine the ETC of a composite material is using numerical simulations with either the finite difference or finite element methods. The numerical simulations are commonly used when the microstructure of the composite is known, but in most cases the analytical models are preferred over the numerical models due to their physical basis, low calculation cost, and reasonable accuracy even when microstructure is uncertain [37]. Even though the numerical models are not as common they can still provide useful information in determining the ETC of composite materials.

2.2.4.1. **Effective Thermal Conductivity of Pebble Bed Reactor Fuel**

A recent study performed by Stainsby et al. [38] investigated the heat transfer in a pebble bed high temperature gas reactor (HTGR). A part of the study involved determining the ETC of the spherical fuel used in the pebble bed reactor. The fuel used in the pebble bed reactor is similar to the TRISO fuel compacts investigated in this study. The pebble bed fuel uses TRISO particles very similar to the particles used in the NGNP fuel. The main difference is in the final shape. The NGNP fuel uses TRISO particles mixed with a graphite matrix that is pressed into a cylindrical compact. The pebble bed fuel presses the TRISO particles and graphite into a sphere of approximately 60 mm in diameter.

Finite Element Analysis (FEA) software was used with three different models to calculate the ETC of the pebble. Three-dimensional models were created based upon the simple cubic (SC), body-centered cubic (BCC), and face-centered cubic (FCC) unit cell structures. The dimensions for each unit cell were determined by the size of the particles and the chosen packing fraction. The representative packing fraction for the pebble bed fuel is 9.344%. The particles were modeled using all the TRISO layers and constituent materials. The particle dimensions and thermal conductivity of the particle constituents can be seen in Table 2.1. The thermal
conductivity of the graphite matrix was 15 W·m⁻¹·K⁻¹. The final geometry used in the FEA study was three unit cells stacked vertically. The boundary conditions included a temperature of 1273 K at the top surface and 773 K at the bottom surface.

Table 2.1. TRISO particle layer dimensions and thermal conductivity.

<table>
<thead>
<tr>
<th>Region</th>
<th>Material</th>
<th>Outer Diameter (m)</th>
<th>Thermal Conductivity (W·m⁻¹·K⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kernel</td>
<td>Uranium Dioxide</td>
<td>500 x 10⁻⁶</td>
<td>3.7</td>
</tr>
<tr>
<td>Coating 1</td>
<td>Porous Pyrolytic Carbon (Buffer)</td>
<td>690 x 10⁻⁶</td>
<td>0.5</td>
</tr>
<tr>
<td>Coating 2</td>
<td>Inner Pyrolytic Carbon</td>
<td>770 x 10⁻⁶</td>
<td>4.0</td>
</tr>
<tr>
<td>Coating 3</td>
<td>Silicon Carbide</td>
<td>840 x 10⁻⁶</td>
<td>16.0</td>
</tr>
<tr>
<td>Coating 4</td>
<td>Outer Pyrolytic Carbon</td>
<td>920 x 10⁻⁶</td>
<td>4.0</td>
</tr>
</tbody>
</table>

The ETC of the models was calculated using the following equation:

$$ k_e = \frac{L q}{\Delta T A_e} $$

(2.28)

where $L$ (m) is the length of the model, $q$ (W) is the total heat flow, $\Delta T$ (K) is the temperature difference, and $A$ (m²) is the cross-sectional area of the model. Equation (2.28) is the same method used to calculate the thermal conductivity for the measurement system, but in the FEA study the heat flow was determined by defining a 2-D horizontal surface in the model and then integrating the heat flux over the surface.

The TRISO particles used for the NGNP fuel and the pebble bed fuel are composites of themselves. In order to compare the FEA results to an analytical ETC model, a thermal conductivity value for the particle ($k_p$) needs to be known. In order to determine a thermal conductivity for the particle, Stainsby applied the same method used to derive Maxwell’s equation (Equation (1.2)). The derivation resulted in twelve unknown coefficients that had to be determined. The report by Stainsby et al. [38] can be referred to for a more detailed explanation of the ETC derivation for the particle. The thermal conductivity of the particle was determined to
be 4.1328 W·m⁻¹·K⁻¹. This value was also verified using the FEA models. The geometry of the particle was changed so that the particle was a homogeneous (smeared) material with a thermal conductivity of 4.1328 W·m⁻¹·K⁻¹ instead of the different layers and materials.

The results from the FEA study show very good agreement between the model with layers and the homogeneous model. The results from Stainsby can be seen in Table 2.2. The results show that the ETC is insensitive to the packing arrangement of the particles. The results also show that a homogeneous particle thermal conductivity of 4.1328 W·m⁻¹·K⁻¹ provides very similar results to the layered particle. The FEA results were also compared to the ETC predicted with Maxwell’s equation (Equation (1.2)). Using a matrix thermal conductivity \(k_m\) of 15 W·m⁻¹·K⁻¹ and a particle thermal conductivity \(k_p\) of 4.1328 W·m⁻¹·K⁻¹ Maxwell’s equation yields an ETC of 13.7 W·m⁻¹·K⁻¹ which results in a maximum difference of 0.36% as compared to the FEA results.

<table>
<thead>
<tr>
<th></th>
<th>SC Smeared</th>
<th>SC Layered</th>
<th>BCC Smeared</th>
<th>BCC Layered</th>
<th>FCC Smeared</th>
<th>FCC Layered</th>
</tr>
</thead>
<tbody>
<tr>
<td>Length (mm)</td>
<td>4.902</td>
<td>4.902</td>
<td>6.1764</td>
<td>6.1764</td>
<td>5.188</td>
<td>5.188</td>
</tr>
<tr>
<td>Cross Sectional Area (mm²)</td>
<td>2.670</td>
<td>2.670</td>
<td>4.239</td>
<td>4.239</td>
<td>6.729</td>
<td>6.729</td>
</tr>
<tr>
<td>(T_f) (K)</td>
<td>1273</td>
<td>1273</td>
<td>1273</td>
<td>1273</td>
<td>1273</td>
<td>1273</td>
</tr>
<tr>
<td>(T_b) (K)</td>
<td>773</td>
<td>773</td>
<td>773</td>
<td>773</td>
<td>773</td>
<td>773</td>
</tr>
<tr>
<td>(\Delta T) (K)</td>
<td>500</td>
<td>500</td>
<td>500</td>
<td>500</td>
<td>500</td>
<td>500</td>
</tr>
<tr>
<td>Heat Flow (W)</td>
<td>3.737</td>
<td>3.738</td>
<td>4.719</td>
<td>4.718</td>
<td>8.926</td>
<td>8.918</td>
</tr>
<tr>
<td>Conductivity ((W·m⁻¹·K⁻¹))</td>
<td>13.72</td>
<td>13.73</td>
<td>13.75</td>
<td>13.75</td>
<td>13.76</td>
<td>73.75</td>
</tr>
</tbody>
</table>

The same type of study done by Stainsby could be done to model the NGNP fuel compacts. The results could be used to compare to the analytical ETC models discussed in Section 2.2.3. The FEA model could be changed to use the current thermal conductivity values of
the NGNP constituent materials. The packing fraction of the model could be changed to represent
the appropriate packing fraction of the NGNP fuels. This information could be used to determine
if there is a more representative analytical ETC model for predicting the fuel compact
conductivity.
CHAPTER 3

OBJECTIVES

The main objective of this work is to characterize the effective thermal conductivity of TRISO fuel compacts. Parts of this work are a continuation of the work performed by Jensen [25]. The main goal of his work was to develop a system that could measure the thermal conductivity of the TRISO fuel compact in a non-destructive manner, over a temperature range of 100°C to 800°C. The goal to characterize the effective thermal conductivity of the TRISO fuel compacts can be separated into three parts.

- Validate/Improve Measurement System
  - Validate the accuracy of the measurement system using certified reference materials and materials of known thermal conductivity
  - Develop a correction scheme as was suggested by the ASTM E 1225 [22] standard
- Sample Measurements
  - Measure the four surrogate TRISO samples supplied by Babcock & Wilcox
  - Measure the six matrix-only samples supplied by Oak Ridge National Laboratory
- TRISO Fuel Thermal Conductivity Modeling
  - Develop unit-cell models of the fuel and perform a FEA study to compare the effective thermal conductivity to models from literature
  - Develop a FEA model with random particle orientations to compare effective thermal conductivity with models from literature
  - Find a model that predicts the effective thermal conductivity of the TRISO fuel accurately
4.1. **Introduction**

In order to understand the scope of this work it is necessary to provide a brief introduction on the work performed by Jensen [25]. The main objectives of his work involved: 1) performing a comprehensive literature review of appropriate methods to measure the thermal conductivity of the TRISO fuel compacts, 2) based upon the literature review a prototype design was constructed, 3) initial testing of the system for calibration, and 4) measure one surrogate TRISO compacts and one AGR-2 matrix-only sample. The samples measured for calibration included Stainless steel 304, Inconel 625, and 99.95% high purity iron. The results obtained showed good agreement with literature values of the samples. Also no correction was applied to the measured data to account for the extraneous heat flow, as suggested by the ASTM E 1225 standard [22].

4.2. **Validation**

The first objective of this work was to validate the thermal conductivity measurement system previously designed. Validation of the system will provide confidence that the results obtained are correct. In order to validate the system, samples of known thermal conductivity must be measured with the system.

4.2.1. **Validation Materials**

The samples chosen to validate the system were chosen to provide a range of expected thermal conductivity values for the TRISO fuel compacts. The expected conductivity range of the TRISO fuel samples is $3 < k < 70 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$. 
4.2.1.1. Pyroceram 9606

For the low thermal conductivity range a glass-ceramic from the Institute for Reference Materials and Measurements (IRMM) [48] was measured. The thermal conductivity varies from 4.06 W·m\(^{-1}\)·K\(^{-1}\) at 298 K to 2.83 W·m\(^{-1}\)·K\(^{-1}\) at 1025 K with a reported uncertainty of ±6.5% [49, 50]. The dimensions of the glass-ceramic sample are 13.8 mm in diameter and 23.3 mm length. These dimensions are slightly different than the dimensions for the TRISO fuel compacts, so the measurement section had to be altered slightly to accept this sample.

4.2.1.2. Inconel 625

The second sample measured was chosen to represent samples in the middle of the expected thermal conductivity range for the fuel compacts. The sample chosen was Inconel 625. The sample was obtained from ESPI metals and was machined to match the dimensions of a fuel compact (12.5 mm diameter x 25.5 mm length). The thermal conductivity values for the Inconel sample were obtained from Maglic [51]. The thermal conductivity varies from 13 W·m\(^{-1}\)·K\(^{-1}\) at 200°C to 21.9 W·m\(^{-1}\)·K\(^{-1}\) at 700°C. These values are more representative of what is expected for the TRISO fuel compacts.

4.2.1.3. 99.95% Pure Iron

The last sample chosen was to represent the upper bounds of the expected conductivity range for the fuel samples. The sample was 99.95% pure iron from ESPI Metals. The dimensions of the iron sample were measured to be 12.8 mm in diameter by 25.5 mm in length. Samples from the remaining stock were prepared and sent to Netzsch for laser flash measurements to obtain reference data. The thermal conductivity values from Netzsch varies from 73.9 W·m\(^{-1}\)·K\(^{-1}\) at 50°C to 34.8 W·m\(^{-1}\)·K\(^{-1}\) at 750°C. The reported uncertainty from Netzsch for the thermal conductivity measurements was ±7%. Literature values for 99.99% pure iron recommended by TPRC [52] were also used for comparison. The thermal conductivity values from TPRC varies from 74.4
W·m⁻¹·K⁻¹ at 77°C to 32.6 W·m⁻¹·K⁻¹ at 727°C. A plot for comparison of the Netzsch and TPRC data can be seen in Figure 4-1. The TPRC and Netzsch data shows very good agreement.

![Pure Iron](image)

Figure 4-1. Comparison between 99.99% pure iron TPRC [52] literature data and 99.95% pure iron samples measured by Netzsch

4.2.2. Testing Procedure

The following will discuss the assembly and measurement procedure for the thermal conductivity measurement system. This explanation will provide a brief description, and a more detailed explanation of the testing procedure is in the work performed by Jensen [25].

4.2.2.1. Assembly Procedure

The assembly of the measurement section for testing is a delicate and time consuming process. The assembly steps are as followed:
1. The sample, top meter bar, and bottom meter bar contact surfaces are polished to provide good contact through the sample column. See Figure 2-3 for a schematic of the measurement section.

2. The length and diameter of each sample is measured with a micrometer.

3. 0.127-mm (0.005”) type N thermocouples insulated with Nextel 312 sleeving are attached to the sample and meter bars. The thermocouples are held in place with 0.254-mm (0.01”) nichrome wire (Figure 4-2).

![Figure 4-2. Type N thermocouples attached to sample using nichrome wire.](image)

4. The distance between the thermocouples for the sample and meter bars is measured using a Canon T1i 15.1 megapixel camera. The pictures are imported into MATLAB and a program is used to measure the fraction of the distance between thermocouples to the overall length of the sample.

5. All the thermocouple beads are coated with Omega CC High Temperature Cement to ensure good contact with the sample and meter bars (Figure 4-3).
6. Once the cement has dried on the sample and meter bars the sample column is assembled.

To reduce the contact resistance a 12.7-μm (0.0005”) piece of nickel foil is placed between the bottom meter bar and sample, and the top meter bar and sample. To ensure alignment of the sample column, a 127-μm (0.005”) nickel foil band is wrapped around the perimeter at the interfaces between the sample and meter bars. The final assembly is shown in Figure 4-4.
Figure 4-4. Assembly of sample column with nickel foil band used to align the column.

7. The guard is then screwed into place over the sample column onto the heat sink. Diatomaceous earth powder is filled in between the guard and sample column for insulation. The sample column heater is then placed into the top meter bar, and a support tube is placed onto the top meter bar. The support tube is used to transfer a constant force...
through the sample column from a spring installed at the top of the support tube. The remaining support columns are installed to secure all the components together.

8. With the measurement section assembled, thermocouples are attached to the guard with the Omega cement. A sheet of insulation is then wrapped around the guard. The completed assembly can be seen in Figure 2-4.

9. The measurement section is then placed into the tube furnace (Figure 2-5). All thermocouple connections are connected to the data acquisition system, and the tube is sealed. The tube is then purged with a vacuum pump and backfilled with ultra-high purity argon.

4.2.2.2. Running Procedure

Once the measurement system is assembled and installed in the furnace the measurement is started. The system is programmed to run at a certain temperature until steady-state conditions are met. According to the ASTM E 1225 standard [22], steady-state conditions are met when the thermocouple readings vary no more than ±0.05 K/hr. Once the steady-state conditions are met the data is taken and the thermal conductivity is calculated for the sample using Equations (2.2) and (2.3). The temperature is then increased 50°C and the procedure is repeated. The system is run up to 700°C and then back down to 100°C alternating between the 50°C increments. The final data has thermal conductivity measurements every 25°C from 100°C to 700°C.

4.2.3. NGNP Sample Information

4.2.3.1. Surrogate Samples

Four surrogate TRISO fuel compacts were sent from Babcock & Wilcox (B&W) to Utah State University for thermal conductivity measurements. The surrogate sample dimensions were ~12.4 mm in diameter and ~30 mm long, a little longer than the standard length of 25 mm for the
fuel compacts. Thus the measurement section was altered to accommodate the additional length of the surrogate sample. The surrogate samples contain zirconium dioxide (ZrO$_2$) as the kernel material with the standard TRISO coatings. The samples were pressed at a targeted packing fraction of 46% and a matrix density goal of 1.75 g/cm$^3$ from lot 13015. The final estimated density and packing fraction were 1.72 g/cm$^3$ and 47.7%, respectively. More information on the surrogate samples can be found in Table 4.1.

Table 4.1. Surrogate sample composition and manufacturing information [53].

<table>
<thead>
<tr>
<th>Lot</th>
<th>Matrix</th>
<th>Temperature (°C)</th>
<th>Hold Time (s)</th>
<th>Pressure (MPa)</th>
<th>Est. Matrix Density (g/cm$^3$)</th>
<th>Est. Packing Fraction (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>13015</td>
<td>Plenco 14838</td>
<td>175</td>
<td>60</td>
<td>18.1</td>
<td>1.720</td>
<td>47.74</td>
</tr>
</tbody>
</table>

4.2.3.2. Matrix-only Samples

Six matrix-only samples were sent from Oak Ridge National Laboratory (ORNL) to Utah State University for thermal conductivity measurements. The matrix-only samples arrived with a ~25 mm diameter and ~60 mm length. They were machined to match the dimensions of the surrogate samples. More information about the matrix-only samples and their constituents is in Table 4.2. The RDKRS samples were from the AGR-2 trial; while the other samples were made using new resins. The new resins are harder than the AGR-2 resin and, as a result, were more brittle [54]. After machining, multiple circumferential cracks were apparent, as seen in Figure 4-5. These cracks were also observed during tests at ORNL [55] so x-rays were taken and the cracks are clearly seen throughout the sample (Figure 4-6). These cracks are expected to have a significant effect on the effective conductivity of the sample. Each crack adds thermal resistance in the sample and since the severity and number of cracks in each sample is unknown the variation in ETC can be large.
Figure 4-5. ARB-B3 sample showing circumferential cracks throughout.

Figure 4-6. X-ray image of ARB-B1 ring blank from ORNL [55].

Table 4.2. Matrix-only sample information [53].

<table>
<thead>
<tr>
<th>Sample ID</th>
<th>Matrix ID</th>
<th>Resin ID</th>
<th>Density (g/cm³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>RDKRS-Z016</td>
<td>RDKRS</td>
<td>Hexion Durite SC-1008</td>
<td>1.6-1.65</td>
</tr>
<tr>
<td>RDKRS-Z050</td>
<td>RDKRS</td>
<td>Hexion Durite SC-1008</td>
<td>1.6-1.65</td>
</tr>
<tr>
<td>ARB-B1-Z008</td>
<td>Blend I</td>
<td>Hexion Durite SD-1708 w/ 5% Hexa</td>
<td>~1.75</td>
</tr>
<tr>
<td>ARB-B1-Z030</td>
<td>Blend I</td>
<td>Hexion Durite SD-1708 w/ 5% Hexa</td>
<td>~1.75</td>
</tr>
<tr>
<td>ARB-B3-S1</td>
<td>Blend III</td>
<td>Plenco 14838 w/ 5% Hexa</td>
<td>~1.75</td>
</tr>
<tr>
<td>ARB-B3-S2</td>
<td>Blend III</td>
<td>Plenco 14838 w/ 5% Hexa</td>
<td>~1.75</td>
</tr>
</tbody>
</table>
4.3. Correction Procedure

As suggested by the ASTM E 1225 standard, a correction should be applied to the thermal conductivity calculations to account for any extraneous heat flow. If the thermal conductivity of the sample and meter bars are not large in comparison to the insulation then radial heat flow into or out of the sample and meter bars is not accounted for in Equations (2.2) and (2.3) (which assume a 1-D heat flow in the meter bars and sample). This can result in large errors in the thermal conductivity calculation. Large systematic error can also occur if the system is poorly designed and ill-operated. The ASTM E 1225 standard suggests that the guard temperature gradient should be either: approximately linear so that the guard temperature matches the temperatures at the two ends of the sample column, or constant with respect to $z$. See Figure 4-7 for a representation of the suggested guard temperature gradients.

![Figure 4-7](image-url)  
Figure 4-7. Representation of the two guard temperature gradients suggested in the ASTM E 1225 standard [22].
Previous work by Xing and Jensen [56] performed a detailed numerical study on the systematic uncertainty of the Guarded Comparative Longitudinal Heat Flow Technique. The study included the effect insulation, geometry, and working conditions (specifically different guard temperature gradients) had on the calculated thermal conductivity error. Interestingly, the study found that the error could be reduced by applying an “optimum guarding” condition instead of the “linear matched” or “constant” guard temperature gradient (see Figure 4-8). Therefore if possible the operating conditions of the system should be controlled to reduce the systematic error. However due to restrictions of the furnace, geometry of the sample and meter bars, insulation, or other parameters of the system it may not be possible to operate using the “optimum guarding” condition. Not operating at the best conditions will induce error into the system; therefore a correction based on the measurement result is necessary to reduce the systematic error of the system.

Figure 4-8. Representation of the "optimum guarding" condition suggested by Xing and Jensen [56].
4.3.1. Correction Scheme Steps

The correction scheme is based on the temperature measurements on the sample column and guard as well as a numerical simulation. The steps are as follows:

1. Design the experimental instruments based on the guidelines from [24–26]. The geometry of the measurement system will be used to create a numerical model. Accurate measurements of the geometry and temperature data from the test are the baseline for the correction.

2. Calculate the test sample thermal conductivity (k_s) using Equations (2.2) and (2.3).

3. Record the temperatures on the two ends of the sample column and the guard temperature distribution. Also record the location for each of these temperatures. These will be used as boundary conditions in the numerical model.

4. Create a numerical model from Step 1 (similar to the model used by Xing and Jensen [56]). Apply the boundary conditions from Step 3. Radial temperature distributions between the ends of the sample column and the guard can be applied based on a 1-D radial heat flow. Apply temperature dependent thermal conductivities to the meter bars, insulation and guard. Apply a guessed conductivity value (at the mean temperature) for the test sample (k_a).

5. Run the model and save the temperatures at the same locations the thermocouples are located on the sample column. These temperatures are used to calculate an output thermal conductivity (k_{so}) from Equations (2.2) and (2.3). This calculated thermal conductivity will account for all the effects due to radial heat flow unlike the value calculated in Step 2.
6. Adjust the guessed thermal conductivity \( (k_{si}) \) and repeat Step 5 until the measured \( (k_s) \) and calculated \( (k_{so}) \) are equal. This step can be simplified by a parametric study on \( k_{si} \) over the range of possible test sample thermal conductivities.

7. The guessed thermal conductivity \( (k_{si}) \) that provides equal measured \( (k_s) \) and calculated \( (k_{so}) \) thermal conductivities is the corrected conductivity value for the sample.

4.3.2. *Insulation Effective Thermal Conductivity Determination*

In Step 4, the temperature dependent material properties of the meter bars and guard are easily determined. The thermal conductivity of the diatomaceous earth powder insulation is difficult to define because it is a combination of the powder and the protective gas. The ETC of a porous material is dependent on the conductivity of the powder, gas, and the packing fraction [57]. At high temperatures the radiation between the particles in the powder also becomes significant. Also in this experiment the protective argon gas is not stationary but flows through the system at a slow, steady rate, thus convection may contribute to the ETC as well.

Due to the complexity of calculating the ETC of the insulation, the value was determined experimentally. To determine the ETC of the insulation a single, solid bar of stainless steel 304 was used in place of the three-piece sample column. The advantage of the solid bar is the elimination of: assembly misalignment, dimension mismatch, contact resistance between the interfaces, and uncertainty from the meter bar material. A series of conductivity measurements on the solid bar was conducted by varying guarding conditions or the specimen mean temperature. The correction scheme was applied to the data from the solid bar test. In the numerical model the insulation thermal conductivity was modified based on a semi-empirical equation to minimize the difference between the measured and calculated thermal conductivity. The modified semi-empirical equation for the insulation ETC is then used in Step 4 for all remaining data corrections.
4.3.3. **Correction Scheme Validation**

To validate the correction scheme a series of tests were conducted with the thermal conductivity measurement system. Tests were performed on three different samples: stainless steel 304, Pyroceram 9606, and 99.95% pure iron.

4.3.3.1. Stainless Steel 304 Correction

Figure 4-9 presents the measured and corrected thermal conductivity of stainless steel 304 (SS 304) at a range of temperatures. Since SS 304 was used to measure SS 304, only a narrow temperature range, ~15 °C was considered. When the guarding condition (guard temperature gradient) was adjusted, the measured value varied significantly. However, the reference conductivity as well as the corrected conductivity value, varies slightly. When different protective gases were employed, the measured conductivity had larger deviations with the higher conductivity gas (helium). If the working condition was not set properly, the measured temperature dependent trends of the thermal conductivity were opposite from the actual one. Since the specimen has the same thermal conductivity as the meter bar, no correction is necessary for the linear matched guarding condition (optimum guarding condition is the same as the linear match because sample and meter bars are the same material).
4.3.3.2. Pyroceram 9606 Correction

Figure 4-10 presents the measured and corrected thermal conductivity of the certified glass-ceramic, Pyroceram 9606. The thermal conductivity of this ceramic is much smaller than the reference meter bar and has an opposite temperature dependent trend from SS 304. Two different assemblies were tested on the same sample but different guarding conditions were employed. The temperature difference between the hot end of the guard and sample column was adjusted slightly in each measurement to maintain a fairly constant ratio of guard temperature difference to sample column temperature difference.

In each measurement, the measured conductivity is fairly constant or at certain ranges increases with temperature. However, the certified value has a definite decreasing trend. However, if the correction scheme is applied, the corrected values from the two measurements all
closely follow the certified curve. The small difference between the two measurements is due to the precision error in the experiments.

At a temperature of ~100°C, the specimen thermal conductivity was measured by varying the guarding condition. Correspondingly, the measured value varied from larger than the certified value to smaller. After correction was made, they were all shifted close to the certified curve. If “optimum guarding” was strictly followed, no correction was necessary for the measured value.

Figure 4-10. Measured and corrected thermal conductivity valued of Pyroceram 9606.

4.3.3.3. 99.95% Pure Iron Correction

In order to validate the correction scheme with a specimen whose thermal conductivity is larger than that of the meter bar, an iron sample was adopted (Figure 4-11). Two sets of assemblies were measured independently and separated by a year span. The first test, using helium as the protective gas, did not apply either the “linear match” or “optimum guarding”
condition while the second test, using argon as the protective gas, moderately followed the “optimum guarding” method. After corrections, both of the two tests rendered very close results and formed one curve. At two temperatures, the thermal conductivity at different guarding conditions was investigated but as can be seen, they all fell on the same curve after correction. When “optimum guarding” was applied, the measured values did not differ much from the corrected ones. If “optimum guarding” is adopted, correction on the measurement is not mandatory, but in the cases when “optimum guarding” is not possible the correction scheme should be applied.

Figure 4-11. Measured and corrected thermal conductivity values of 99.95% pure iron.

4.4. Numerical Study

Based upon the success of the FEA study performed by Stainsby et al. [38] to model the ETC of the pebble bed fuel, the goal was to do a similar study using more appropriate values
characteristic of the NGNP TRISO fuel. The study performed by Stainsby was for a single packing fraction and a single matrix thermal conductivity. This work will explore the effects of higher packing fractions, a range of matrix thermal conductivity, and also applying a radioactive heat source to the TRISO particle kernel. The results can then be compared to the analytical effective thermal conductivity models discussed in Section 2.2.3. This will reveal which ETC model predicts the conductivity of the TRISO fuel compacts best. This model can then be used in place of the German correction factor (Equation (1.1)) in the performance and safety modeling of the fuel.

4.4.1. Unit Cell Model

4.4.1.1. Method

Geometry

The numerical calculations were performed using COMSOL Multiphysics. Three different geometries were made based on: simple cubic, body-centered cubic, and face-centered cubic unit cells. The three different geometries could be compared to see if the packing arrangement would have an effect on the ETC. The three different models can be seen in Figure 4-12 through Figure 4-14.

To study the effect of varying packing fractions, the model geometry was made parametric so that all the dimensions changed according to the desired packing fraction. Looking at the results for a range of particle packing fraction helps in determining which analytical ETC model provides the most accurate results for packing fractions expected for the TRISO fuel compacts. For this work the studied packing fractions were between 20-40%. Figure 4-15 shows the BCC geometry at 20% and 40% packing fractions.
Figure 4-12. Simple cubic (SC) geometry.

Figure 4-13. Body-centered cubic (BCC) geometry.
Figure 4-14. Face-centered cubic (FCC) geometry.

Figure 4-15. BCC model with 20% and 40% packing fractions, respectively.

“Homogeneous” Particle Thermal Conductivity

Because the analytical models of ETC in Section 2.2.3 are for two phase composites, models with non-layered (homogeneous) particles were created so that the homogeneous thermal conductivity of 4.1328 W·m⁻¹·K⁻¹ determined by Stainsby et al. [38] could be validated. Figure 4-16 shows the SC model with homogeneous particles and TRISO particles.
Figure 4-16. Homogeneous model and layered model, respectively.

For all the models with layered particles the dimensions of the particle are the same, as well as the thermal conductivity of the different materials. The dimensions for the different layers were determined from multiple resources [38, 59, 60]. The thermal conductivity for the different layer materials was also determined from multiple resources [3, 38, 59]. The values for both can be seen in Table 2.1. If there were any discrepancies between the different sources then the values from Stainsby et al. [38] were used so that the results could be compared with the results obtained by Stainsby. This allowed a check for the accuracy of the model. The diameter for the homogeneous particle was chosen to be the same as the outer diameter of the last coating in the TRISO particle. The thermal conductivity was chosen to be 4.1328 W·m⁻¹·K⁻¹ as found by Stainsby.

Radioactive Kernel

The surrogate samples received from B&W that were tested are not made with a radioactive fuel kernel. In reality the particles will have a radioactive kernel that is producing a large amount of heat. The addition of a radioactive kernel could have an effect on the ETC of the TRISO fuel compact. In order to determine the magnitude of the effect, the model was solved
with a kernel generating heat and also with no heat generation. According to Stainsby et al. [38] the power density for a single particle is 50 W/mm$^3$. That value was used to determine the volumetric heat generation of the kernel to be approximately $3.2 \times 10^8$ W/m$^3$. That value was applied to just the kernel in the COMSOL model (Figure 4-17).

![Figure 4-17](image)

Figure 4-17. Applying a heat generation source to the kernel of the TRISO particle.

4.4.1.2. Theory

The geometry used for the numerical model was different than what was presented in Figure 4-12 through Figure 4-14. Five unit cells in the direction of heat flow were used as the geometry (Figure 4-17). This was done so that values could be taken from the middle unit cell where the temperature gradients and heat flux was not affected by the boundary conditions applied to the top and bottom of the model. This geometry was also used so that a direct comparison could be made to the results obtained by Stainsby et al. [38]. A temperature was
specified at the top surface and a heat flux was applied to the bottom surface. The temperature applied to the top was 373.15 K, which is really an arbitrary number because in this model none of the properties are temperature dependent. A heat flux was applied so that the heat rate through the unit cell was 0.02 W. This value was chosen because it provided a temperature gradient through the sample similar to temperature gradients measured in the surrogate TRISO samples. Symmetry boundary conditions were applied to the four sides parallel to the heat flow. Figure 4-18 shows the BCC model that was used in the COMSOL simulations with the boundary conditions.

![COMSOL model and boundary conditions](image)

Figure 4-18. COMSOL model and boundary conditions.

The ETC is calculated using the same method as the guarded-comparative-longitudinal heat flow method. The equation used to calculate the ETC is Equation (2.3). The heat flow is determined using a surface integral of the heat flux in COMSOL at the top and bottom surfaces of
the center unit cell (Figure 4-19). Just as the heat flow \((q_x)\) in Equation (2.3) is the average of the heat flow in the two meter bars, the COMSOL model also averages the heat flow from the two surfaces. This average does not matter when there is not heat generation in the fuel kernels because the heat rate is the same at the two surfaces, but when there is heat generation in the kernel then the heat flow is not the same on the two surfaces and so the average is used.

![Top and Bottom of Center Unit Cell](image.png)

Figure 4-19. Locations where heat flow and temperature are calculated.

The term \(\Delta T_x\) in Equation (2.3) is the temperature difference between the top and bottom surfaces of the center unit cell. An average temperature at each surface was found using COMSOL and the difference was taken to get the \(\Delta T_x\) term. The \(\Delta Z_x\) term is just the height of the unit cell, which is a function of the packing factor. \(A_x\) is the cross-sectional area of the unit cell that is perpendicular to the heat flow.
In addition to using Equations (2.2) and (2.3) to determine the ETC of the numerical model, another method was used to determine the ETC. The method was based upon solving the heat equation and then solving for the thermal conductivity based upon the boundary conditions from the numerical model.

Starting with a generic form of the heat equation

\[ \frac{\partial}{\partial x} \left( k \frac{\partial T}{\partial x} \right) + \frac{\partial}{\partial y} \left( k \frac{\partial T}{\partial y} \right) + \frac{\partial}{\partial z} \left( k \frac{\partial T}{\partial z} \right) + \dot{q} = \rho c_p \frac{\partial T}{\partial t} \]  

(4.1)

where \( \dot{q} \) is the volumetric heat generation term, \( \rho \) is the density, \( c_p \) is the specific heat, and \( k \) is the thermal conductivity. The numerical model can be approximated as a 1-D equation; it is also a steady-state problem and the thermal conductivity is a constant so Equation (4.1) can be simplified down to

\[ \frac{\partial^2 T}{\partial z^2} + \frac{\dot{q}}{k} = 0. \]  

(4.2)

Solving Equation (4.2) results in

\[ T(z) = -\frac{\dot{q}}{2k} z^2 + C_1 z + C_2. \]  

(4.3)

Using the COMSOL solution, multiple boundary conditions can be applied to Equation (4.3) to determine the unknown constants. A heat flux boundary condition at \( z = 0 \) (bottom of unit cell), and a temperature boundary was applied at \( z = L \) (where \( L \) is the height of the unit cell). The two boundary conditions are:

\[ -k \frac{dT(0)}{dz} = q'' \]  

(4.4)

\[ T(L) = T_t \]  

(4.5)

where \( q'' \) is the heat flux and \( T_t \) is the temperature at the top of the unit cell. Applying Equations (4.4) and (4.5) to Equation (4.3) results in
The ETC of the unit cell based upon the boundary conditions from the numerical model is determined by solving Equation (4.6) for \( k \) with the following condition

\[
T(0) = T_b
\]

where \( T_b \) is the temperature at the bottom of the unit cell. Applying Equation (4.7) to Equation (4.6) the ETC is:

\[
k_e = \frac{L(L\dot{q} + 2q^*)}{2(T_b - T_i)}
\]

In the cases where there is no heat generation (\( \dot{q} \)) term then Equation (4.8) simplifies into Equation (2.3).

4.4.2. Random Model

The FEA models based upon the three different unit cells will provide valuable information to the study. In reality the particle arrangement in the TRISO fuel compact is not an ordered structure. The particle arrangement will be random. Also many of the analytical effective conductivity models assume a random orientation of particles (Geometric Mean model (Section 2.2.3.2), EMT model (Section 2.2.3.5), Lewis and Nielsen model (Section 2.2.3.7), and many other models). In order to develop the most accurate model of the TRISO fuel compacts, a model based upon random particles was developed.

4.4.2.1. Random Particle Generation

A code was written using a computer algebra software, Mathematica, to generate the random positions of the particles. The size of the geometry was based upon the desired packing fraction and some predefined numbers. The geometry was based upon a rectangular prism (square cross section) where the height was three times the length of the sides. The number of particles in
the model was chosen to be 100 particles inside the rectangular prism, and then 10 particles would be located on each of the four sides, and 5 particles would be on the top and bottom. The particles located on the sides, top and bottom would only contribute to half a particle to the model. So in all there were 125 total particles in the model. This number was then used to determine what the height of the prism should be for a certain packing fraction. The equation to determine the size of the rectangular prism was

$$h_r = \left( \frac{1500 \pi r^3}{\phi} \right)^{\frac{1}{3}}$$  \hspace{0.5cm} (4.9)

where $h_r$ is the height, $r$ is the radius of the particle, and $\phi$ is the desired packing fraction. The length of each side ($s$) of the rectangular prism would be $s = h/3$. A representation of the geometry is shown in Figure 4-20.

Figure 4-20. FEA model with random particles positions, showing the half particles on the surface and the particles on the inside of the model, respectively (18% packing fraction).

The Mathematica code first generated the points for the 100 interior particles. The random position for the particle was determined using a random number function. The function
was specified to choose a random number in a certain range. The range for the \( x \) and \( y \) position was between \( r \) and \( s - r \). This ensured that the entire particle would be located within the rectangular prism. The range for the \( z \) position was between \( r \) and \( h_r - r \). As each new random position \((x', y', z')\) for a particle was found it was checked with all the random positions already determined. It was checked to make sure that the distance between each new random position is at least \( 2r \) from any other particle, to ensure that no particles overlap. If the new particle position overlapped with a previous particle the point was rejected. Once all the 100 interior points were generated then the points on the boundary were created. The process for creating the points on the sides and top was the same as the previous except that either the \( x \), \( y \), or \( z \) location was fixed at the boundary. In the end the model consisted of 150 total points, which is equivalent to 125 total spheres within the boundary of the rectangular prism.

The random particle code worked really well for packing fraction of about 35% and under. If higher packing fractions were needed the code had to be modified to accomplish this. The reason for the difficulty in generating points for high packing fractions has to do with the random nature of the code. The distance between two random points could be just far enough apart to not allow another particle to be placed in between, which leaves a gap that is too large for a high packing fraction structure. In reality the particles would be able to move around as the compact is pressed, so those gaps could be filled. The code was changed so that any number of particles could be placed inside the prism. The code used an iteration count to determine how many times a random position was rejected because it conflicted with another particle already in place. Once the iteration count reached a certain number of attempts the number of particles already placed inside the prism was recorded. The code then proceeded to each of the sides, top, and bottom just as the previous code but using the iteration count to achieve as many particles on each of those surfaces as possible. This method resulted in a 39.8% packing fraction where the
previous code could only approach 35%. For comparison refer to Figure 4-21, which has a 39.8% packing fraction, and compare it to Figure 4-20, which has an 18% packing fraction. The model with 39.8% packing fraction has only three particles on the top surface, but it has eleven particles on the two sides shown. The 18% model has five particles on the top and ten particles on the two sides.

Figure 4-21. FEA model with random particles (39.8% packing fraction).

4.4.2.2. COMSOL Multiphysics LiveLink for MATLAB

COMSOL Multiphysics has a feature that links its usage with MATLAB. Scripts can be run in MATLAB that will perform the same operations as using the COMSOL interface. This becomes a very valuable tool when creating a few hundred particles at random locations, and performing the tedious Boolean operations to create the final geometry. This LiveLink for MATLAB was used extensively to create the geometries for the random model.

The LiveLink for MATLAB works the same as the standard features for MATLAB. Individual functions can be sent to COMSOL using the command window, or a list of functions
to be performed can be written in an “m file” and run in the command window. For this work the “m file” approach was used to create the geometry.

4.4.2.3. Geometry Generation

Again Mathematica was used to generate the output used for the “m file”. A program was written to generate a text output of the multiple commands MATLAB would use to create each of the particles at the random locations already generated and perform the individual Boolean operations for each particle. The commands were then imported into COMSOL to create the geometry for the FEA model. The geometry was then exported as a COMSOL geometry file. This was done so that the geometry could be imported into a COMSOL file that had the heat transfer module, all the material properties, boundary conditions, and post processing features already saved. This saved time and only required the geometry file for different packing fractions to be imported and then the model could be solved.

4.4.2.4. COMSOL Operations

The method used to solve the random model was very similar to the method used to solve the unit cell models. In the random model a homogeneous particle was used, and as will be shown later in this document (Figure 6-8, Section 6.2.1), the “homogenized” thermal conductivity of 4.1328 W·m⁻¹·K⁻¹ provides very accurate results to the models with layered particles. So the particles were assigned a thermal conductivity of 4.1328 W·m⁻¹·K⁻¹. The boundary condition at the top surface was a temperature boundary condition of 373.15 K, and the bottom surface had a heat flux boundary condition. In the tests where the particles had a radioactive heat source, the heat generation term was modified so that the same amount of heat generated in the kernel of the layered model would be generated in the homogeneous particle. The heat flow was measured at the top and bottom surface of the rectangular prism using COMSOL. The temperature of the
bottom surface was also calculated in COMSOL. These measurements were used to calculate the ETC using Equation (4.8).
CHAPTER 5
UNCERTAINTY ANALYSIS

For this work a Taylor Series Method (TSM) for propagation of uncertainty was used to analyze the uncertainty in the sample thermal conductivity. The methodology and notation given by Coleman and Steele [61] were used for the uncertainty analysis. As part of this methodology the systematic and random uncertainties were analyzed independently for Equations (2.2) and (2.3).

The measurement of distance between thermocouple locations, $\Delta Z$, has systematic uncertainty from the micrometer and caliper that were used to measure the overall length of the sample and meter bars. The manufacturers of the micrometer and caliper give an uncertainty of 1.6 $\mu$m and 20 $\mu$m, respectively, at the 95% confidence level. The random uncertainties come from three sources:

1. Resolution of the 15.6 megapixel camera. The camera is used to measure the thermocouple locations by comparing the ratio of the thermocouple locations to the overall length of the sample or meter bar. For a 25 mm long test sample the random uncertainty at a 95% confidence level of 5.6 $\mu$m, and 17 $\mu$m for a 75 mm long meter bar.

2. The resolutions of the micrometer and caliper are 1 $\mu$m and 12 $\mu$m respectively, at a 95% confidence level.

3. The size of the thermocouple bead (twice the diameter of the thermocouple wire) of 0.254 mm at the 95% confidence level.

The largest contributor to the random uncertainty in $\Delta Z$ is the size of the thermocouple bead and therefore the other values can be ignored.
The systematic uncertainty in the temperature was taken as 0.15°C at the 95% confidence level; this was suggested by Sweet [20]. This value is much lower than the 2.2°C or 0.75 %, whichever is greater, as recommended by Omega [62] for Type N thermocouples. The reason for the suggestion by Sweet is because: (1) modest temperature differences are being measured, (2) thermocouple wire is taken from the same spool, and (3) the same reference junction and readout devices are being used for all thermocouples. A test was performed to provide evidence of the reduced uncertainty, the results can be seen in the work by Jensen [26]. During the measurement, steady-state conditions are considered met when at least one hour of data has a standard deviation of less than 0.035 K. This value is negligible compared to the systematic uncertainty and will be ignored because the contribution to the random uncertainty is the standard deviation of N (> 360) measurements divided by the square root of the number of measurements.

The 95% systematic uncertainty of the meter bar thermal conductivity is recommended by several sources [20, 58] as 5%. There is no random uncertainty associated with the meter bar thermal conductivity because the values are read from a table. Sweet [20] also suggested that a 1% random uncertainty should be added to account for any non-symmetric heat flow or small details related to the setup. Table 5.1 shows a summary of the systematic standard and random standard uncertainties for the measured variables in Equations (2.2) and (2.3). And Table 5.2 shows sample values from a representative measurement.

Another concern for the uncertainty of the measurement is thermocouple decalibration. At elevated temperatures the small diameter of the thermocouple wires makes them vulnerable to decalibration compared to larger diameters [62]. Decalibration of the thermocouples results in a hysteresis type effect on the measurements as can be seen in the results from Test 1 in Figure 6-1 where the measurements were taken every 50°C up to 550°C, and then back down alternating between the previous measurements. Type S or R thermocouples could be used reduce the effect of decalibration, but due to their increased cost it was not feasible to use them. Also larger
thermocouples could be used, but the larger the thermocouple wires the larger the uncertainty due to the size of the bead (discussed previously). Therefore the decalibration of the thermocouples was closely monitored and the test was terminated when decalibration was noticed.

Table 5.1. Standard uncertainties for measured variables.

<table>
<thead>
<tr>
<th>Variable</th>
<th>Systematic Uncertainty, $b_i$</th>
<th>Random Uncertainty, $s_i$</th>
<th>Units</th>
</tr>
</thead>
<tbody>
<tr>
<td>$k_m$</td>
<td>5% $k_m$ (0.48)</td>
<td>---</td>
<td>W·m$^{-1}$·K$^{-1}$</td>
</tr>
<tr>
<td>$\Delta T_m$</td>
<td>0.075</td>
<td>0.002</td>
<td>°C</td>
</tr>
<tr>
<td>$\Delta Z_m$</td>
<td>20</td>
<td>128</td>
<td>μm</td>
</tr>
<tr>
<td>$q_m$ (nonuniformity)</td>
<td>---</td>
<td>1% $q_m$ (0.011)</td>
<td>W</td>
</tr>
<tr>
<td>$q_s$</td>
<td>0.029</td>
<td>0.012</td>
<td>W</td>
</tr>
<tr>
<td>$\Delta T_s$</td>
<td>0.075</td>
<td>0.002</td>
<td>°C</td>
</tr>
<tr>
<td>$\Delta Z_s$</td>
<td>0.8</td>
<td>127</td>
<td>μm</td>
</tr>
</tbody>
</table>

Table 5.2. Nominal values for measured variables from a sample measurement.

<table>
<thead>
<tr>
<th>Measured Variable</th>
<th>Value</th>
<th>Units</th>
</tr>
</thead>
<tbody>
<tr>
<td>$k_m$</td>
<td>19</td>
<td>W·m$^{-1}$·K$^{-1}$</td>
</tr>
<tr>
<td>$\Delta T_m$</td>
<td>10</td>
<td>°C</td>
</tr>
<tr>
<td>$\Delta Z_m$</td>
<td>20</td>
<td>mm</td>
</tr>
<tr>
<td>$q_m$</td>
<td>1.14</td>
<td>W</td>
</tr>
<tr>
<td>$\Delta T_s$</td>
<td>10</td>
<td>°C</td>
</tr>
<tr>
<td>$\Delta Z_s$</td>
<td>20</td>
<td>mm</td>
</tr>
<tr>
<td>$k_s$</td>
<td>19</td>
<td>W·m$^{-1}$·K$^{-1}$</td>
</tr>
</tbody>
</table>
Using the TSM approach, the systematic standard uncertainty for \( q_s \) is:

\[
b_{q_s}^2 = \left( \frac{\partial q_s}{\partial k_m} \right)^2 b_{k_m}^2 + \left( \frac{\partial q_s}{\partial \Delta T_{m1}} \right)^2 b_{\Delta T_{m1}}^2 + \left( \frac{\partial q_s}{\partial \Delta Z_{m1}} \right)^2 b_{\Delta Z_{m1}}^2 + \left( \frac{\partial q_s}{\partial k_m} \right)^2 b_{k_m}^2 + \left( \frac{\partial q_s}{\partial k_m} \right)^2 b_{k_m}^2 + 2 \left( \frac{\partial q_s}{\partial k_m} \right) \frac{\partial q_s}{\partial k_m} b_{k_m} b_{k_m}.
\]  

(5.1)

Since the nominal values for the measured parameters and uncertainties are equal between meter bars, the subscript \( m \) will replace subscripts 1 and 2 in Equation (5.1). The correlated uncertainties are from the meter bar thermal conductivity which is the same for each meter bar. Therefore Equation (5.1) can be simplified to

\[
b_{q_s}^2 = 4 \left( \frac{\partial q_s}{\partial k_m} \right)^2 b_{k_m}^2 + 2 \left( \frac{\partial q_s}{\partial \Delta T_{m}} \right)^2 b_{\Delta T_{m}}^2 + 2 \left( \frac{\partial q_s}{\partial \Delta Z_{m}} \right)^2 b_{\Delta Z_{m}}^2.
\]  

(5.2)

For the random uncertainties the same arguments can be made as for the systematic uncertainty except that there are no correlated terms. There is also the addition of the nonuniformity in heat flow, \( s_{q_s} \). Thus the random standard uncertainty for the measured heat flow is

\[
s_{q_s}^2 = 2 \left( \frac{\partial q_s}{\partial k_m} \right)^2 s_{k_m}^2 + 2 \left( \frac{\partial q_s}{\partial \Delta T_{m}} \right)^2 s_{\Delta T_{m}}^2 + 2 \left( \frac{\partial q_s}{\partial \Delta Z_{m}} \right)^2 s_{\Delta Z_{m}}^2 + s_{q_s}^2.
\]  

(5.3)

The systematic and random uncertainty equations for the measured sample thermal conductivity can be found by applying the same principles used to derive Equations (5.2) and (5.3). The equations are:

\[
b_{k_s}^2 = \left( \frac{\partial k_s}{\partial q_s} \right)^2 b_{q_s}^2 + \left( \frac{\partial k_s}{\partial \Delta T_{s}} \right)^2 b_{\Delta T_{s}}^2 + \left( \frac{\partial k_s}{\partial \Delta Z_{s}} \right)^2 b_{\Delta Z_{s}}^2.
\]  

(5.4)

\[
s_{k_s}^2 = \left( \frac{\partial k_s}{\partial q_s} \right)^2 s_{q_s}^2 + \left( \frac{\partial k_s}{\partial \Delta T_{s}} \right)^2 s_{\Delta T_{s}}^2 + \left( \frac{\partial k_s}{\partial \Delta Z_{s}} \right)^2 s_{\Delta Z_{s}}^2.
\]  

(5.5)
Using the values from Table 5.1 and Table 5.2 the systematic and random standard uncertainties are:

\[ b_{k_s} = 0.506 \text{ W/m/K}, \]  
\[ s_{k_s} = 0.241 \text{ W/m/K}. \]  

The overall uncertainty for a 95% confidence interval is:

\[ U_{k_s} = 2(b_{k_s}^2 + s_{k_s}^2)^{1/2} = 1.121 \text{ W/m/K}, \]  
\[ \frac{U_{k_s}}{k_s} = 5.9\%. \]  

Removing the uncertainty contribution from the meter bar thermal conductivity, which is the largest contributor, results in an uncertainty of:

\[ \frac{U_{k_s}}{k_s} = 3.1\%. \]  

The overall uncertainty of the measurement system can be assumed to be approximately 6%. Half of the uncertainty in the system comes from the meter bars; therefore it is important to reduce the uncertainty of the meter bars as much as possible.

Note: for a more detailed description of the uncertainty analysis for the measurement system, refer to the work done by Jensen [25, 26] and Xing et al. [56].
CHAPTER 6
RESULTS AND DISCUSSION

6.1. Experimental

The experimental results will be presented below. First the validation results will be presented to instill confidence in the thermal conductivity measurement system’s ability to accurately measure the thermal conductivity of the samples. Then the results of the surrogate samples will be discussed followed by the matrix-only sample results. Finally a comparison of the matrix-only and surrogate results to the German data will be presented. All of the results shown below will represent the corrected thermal conductivity values using the correction scheme presented in Section 4.3.

6.1.1. Validation Results

6.1.1.1. Pyroceram 9606

The results from two separate tests can be seen in Figure 6-1. As can be seen from the results the measured data is well within the 6.5% uncertainty range for the material. The maximum deviation of the two tests from the certified value was under 3%. The results also show how repeatable the measurement system can be. The results show that accurate measurements can be obtained for samples whose thermal conductivity is at the low end of what would be expected from the fuel compacts.
6.1.1.2. Inconel 625

The results for Inconel 625 are shown in Figure 6-2. The maximum deviation from the reported values from Maglic [51] is 6%. The thermal conductivity range of the Inconel is closer to what is expected for the TRISO fuel compacts than the Pyroceram or iron sample.

6.1.1.3. 99.95% Pure Iron

The results can be seen in Figure 6-3. The maximum difference between the two results was 8%. The difference is slightly out of the uncertainty range (±7%) Netzsch provided for their results, but the uncertainty for the measurement system is 6%. It is unsure why there is a bias between the measured and literature values, but the thermal conductivity of the iron is much higher than what will expected for the TRISO fuel compacts.
Figure 6-2. Validation results for Inconel 625 [51].

Figure 6-3. Validation results for 99.95% pure iron compared to recommended TPRC [52] literature values and measurements by Netzsch from the same stock of material.
As can be seen from Figure 6-1 through Figure 6-3 the measurement system results are very close to all the reference materials over the entire expected thermal conductivity range. Therefore confidence can be taken in the results obtained for the fuel compacts using the measurement system.

6.1.2. Surrogate Sample Results

The four surrogate TRISO samples from B&W were the first samples measured. The results for these samples are presented in Figure 6-4. Little information was provided about the surrogate samples (see Section 4.2.3.1). The information received about the surrogate samples (Table 4.1) stated that the matrix material was Plenco 14838, but according to the information about the matrix-only samples (Table 4.2) the name of the resin is Plenco 14838. Thus Plenco 14838 is not the type of matrix material used; therefore it is not known what the matrix material used in the surrogate samples is. Also the Plenco 14838 was one of the new resins that cured harder, which could have caused cracks in the samples that were not visible on the surface. The presence of cracks could explain the large variation between the four samples.

In general the results of the surrogate samples agree well with each other. At low temperatures (~100°C) the maximum variation between the samples was approximately 15 W·m⁻¹·K⁻¹, while at high temperatures (~600°C) the maximum variation was around 10 W·m⁻¹·K⁻¹. The reason for the variation between samples is not precisely known, although, the possible presence of cracks in the matrix or other variations in the exact composition and structure of the composite material will certainly cause some variation. It should be also noted that the values presented here don’t necessarily represent the thermal conductivity values for real TRISO fuel compacts. The samples had a 47.74% packing fraction which is much higher than what is expected (0.3 to 0.38) [25]. Because of the lower ETC of the TRISO particles (~4.1328 W·m⁻¹·K⁻¹), increased packing fraction is expected to have an adverse effect on the ETC. The matrix material is not known for
the samples and it therefore makes an unknown contribution to the ETC. Thus the results for the surrogate compacts provide an estimate of the variation that can exist in a batch of compacts.

![Graph](image)

**Figure 6-4.** Results for surrogate samples from B&W.

6.1.3. *Matrix-only Sample Results*

After the surrogate samples were measured, the six matrix-only samples from ORNL were measured. The results from the measurements can be seen in Figure 6-5. The RDKRS samples from the AGR-2 test show good agreement between samples, while the variation between the other samples was much larger. The variations between the two different ARB samples is attributed to the cracks (see Section 4.2.3.2). There also seems to be a difference between the various matrix blends. The RDKRS samples’ thermal conductivity was on average at least 15% higher than the highest ARB sample.
Figure 6-5. Results for matrix-only samples from ORNL.

When comparing Figure 6-4 and Figure 6-5 it is interesting to note that the thermal conductivity of the surrogate TRISO samples is higher than all the matrix-only samples. According to Maxwell’s Equation (Equation (2.11)) the ETC cannot be higher than the matrix thermal conductivity, unless the particles have a higher thermal conductivity and in this case the inverted form of Maxwell’s Equation is used. For the case of TRISO fuel, all the constituent materials of the TRISO particle, except for silicon carbide, have a thermal conductivity much lower than the matrix. Therefore the larger thermal conductivity of the composite samples is not expected. Explanations for the observed behavior are: 1) the matrix material in the surrogate TRISO samples is not the same as the matrix-only samples (although the matrix conductivity would have to be very large); 2) The addition of the particles in the matrix may induce order into the bonding nature of the typically amorphous graphite material. Such order could create good
conduction paths through the compacts. Even with the same composition, the matrix conductivity is also expected to increase with higher compaction pressure or sintering temperature. To date, limited information regarding the samples prevents a more clear explanation for the observed behavior.

6.1.4. German Comparison

One of the main objectives in this work is to compare the thermal conductivity of the NGNP materials to data reported for German TRISO materials [3, 4]. Because the PARFUME modeling is using the German data (no other data is available), it is important to know if those values correspond well to the NGNP fuel. The German data is plotted with the matrix-only samples in Figure 6-6. The data being used in the model is the German A3-27 data for the matrix thermal conductivity. As can be seen the German data has a much higher thermal conductivity than the matrix-only samples tested, on average it is 57% larger than the RDKRS-Z016 sample.

In order to account for the particles, the PARFUME model multiplies the matrix thermal conductivity values by a correction factor (Equation (1.1)). This correction factor assumes that the thermal conductivity of the particles is zero. Figure 6-7 shows a comparison between the surrogate TRISO samples and the German data with the correction factor applied using the same packing fraction as the surrogate samples. The resulting ETC for the German data is much less than the surrogate samples. On average the German data is 59% less than the 13015B sample and 48% less than the 13015C sample. So the German data does not represent the materials that are currently being produced for the NGNP project. This means that the performance and safety modeling being done is not accurately predicting the temperatures of the fuel inside the reactor.
Figure 6-6. Matrix-only samples compared to German data.

Figure 6-7. Surrogate samples compared to German matrix with a 47.7% packing fraction.
6.2. Numerical

The numerical results will now be presented and discussed. First the validation of the “homogenized” particle thermal conductivity will be presented, followed by a validation of the numerical model. Then the effective conductivity of the numerical study will be presented and compared to the analytical effective conductivity models.

6.2.1. “Homogenized” Particle Thermal Conductivity Validation

Results were obtained for all three unit cell types: SC, BCC, and FCC. Three different packing fractions were used: 20, 30, and 40%. And the matrix thermal conductivity values were changed from 15-60 W·m⁻¹·K⁻¹. These results provided a good enough scope to ascertain confidence that the value of 4.1328 W·m⁻¹·K⁻¹ is a good value to use as the homogeneous thermal conductivity value of the particle. The results in Figure 6-8 show that there is very good agreement between the layered model and the homogeneous model. The largest difference between the ETC of the layered and homogenous model was only 0.5%. It can be concluded that the homogeneous thermal conductivity particle value suggested by Stainsby works well for this model.

![Image](image-url)

Figure 6-8. Validation results for the "homogenized" particle thermal conductivity.
6.2.2. **Numerical Model Validation**

Results from the COMSOL model were validated against the results obtained by Stainsby et al. [38] to ensure that the model was correct. In order to validate the COMSOL model the same geometry and boundary conditions were applied as used by Stainsby et al. [38]. The results in Table 6.1 show that there is a good agreement between the two studies. The largest difference was only 0.335%. This provided confidence in the numerical model for the future calculations.

Table 6.1. Validation results of the COMSOL model to the Stainsby model.

<table>
<thead>
<tr>
<th>Unit Cell Type</th>
<th>COMSOL Conductivity (W·m⁻¹·K⁻¹)</th>
<th>Stainsby Conductivity (W·m⁻¹·K⁻¹)</th>
<th>% Difference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Simple Cubic</td>
<td>13.703</td>
<td>13.73</td>
<td>0.196</td>
</tr>
<tr>
<td>Body Centered Cubic</td>
<td>13.707</td>
<td>13.75</td>
<td>0.310</td>
</tr>
<tr>
<td>Face Centered Cubic</td>
<td>13.704</td>
<td>13.75</td>
<td>0.335</td>
</tr>
</tbody>
</table>

6.2.3. **Numerical Effective Thermal Conductivity Results**

In a numerical study the scope can be very large, especially when the number of variables is large. With this model there were five variables that could be changed, they were:

1. The packing type (SC, BCC, FCC, and random)

2. The packing fraction

3. The matrix thermal conductivity

4. Particles with or without heat generation

5. Layered or homogeneous particles.
Since it had already been shown that a value of 4.1328 W·m$^{-1}$·K$^{-1}$ provided accurate results for a homogeneous thermal conductivity of the particle this part was left out of the effective conductivity comparison study. The remaining four variables were included in the study, but some of them were limited in scope to match the expected characteristics of the TRISO fuel compacts. For example the packing fraction was limited to a study of 20%, 30%, and 40%. The matrix thermal conductivity was also limited to values between 10-60 W·m$^{-1}$·K$^{-1}$. Results were obtained for all four packing types to see if there was any influence of the packing structure on the ETC of the compact. Results were also obtained for heat generation in the particles as well as no heat generation to see if the ETC was affected by the discrete locations of heat generation.

The ETC from the FEA study was compared to multiple analytical effective conductivity models. The results were compared to all of the models discussed in Sections 2.2. Only the most applicable models and the models that showed the most promise were shown in the graphs. The models shown in the graph were the full version of Maxwell’s equation, the reduced version of Maxwell’s equation (German equation), the EMT model, and the Chiew & Glandt model. The remaining models did not provide good estimations for the ETC of the TRISO fuel compacts. It should also be noted that the reduced Maxwell equation did not estimate the ETC of the fuel well but it was included in the graphs to provide comparison to the German equation.

The results are presented in Figure 6-9 through Figure 6-11 for different thermal conductivity ratios ($\kappa = k_p/k_m$) to avoid too much information on a single plot. As can be seen the three different unit cell packing types all provide very close results to each other. This seems to show that the arrangement of particles has very little effect on the ETC. The models based upon the unit cells agree best with the full Maxwell equation over the entire range of packing fractions, even though it was stated that the Maxwell equation is only valid for very low packing fractions [32, 63]. On average the reduced Maxwell equation shows the largest deviation from the numerical results. A surprising result was how the EMT model deviated from the numerical
results more as the packing fraction increased. The EMT model is supposed to be more accurate for larger packing fractions ($\phi << 0$). One explanation could be that the EMT model assumes a random distribution of particles and in the numerical results the particles are not in a random distribution.

The results from the random model show very good agreement with the Chiew & Glandt model. This was expected since the Chiew & Glandt model is an improved version of the Maxwell equation. It was also very interesting that the random results did not agree with the EMT model. The EMT model was derived based on a random distribution and for higher packing fractions. The EMT model also assumes no distinction between the continuous phase and the dispersed phase, which in the case of the TRISO fuel compacts is not an accurate assumption.

The repeatability of the random model was tested to see the deviations between models with the same packing fraction. Multiple tests were run at a packing fraction of 30% and 35% with a different random model each time. As can be seen in Figure 6-9 through Figure 6-11 there is a small variation between tests, but the average standard deviation of the tests at the same packing fraction was less than 0.08 W·m$^{-1}$·K$^{-1}$.

Another method of looking at the results is to plot the results as a function of thermal conductivity ratio ($\kappa$) at different packing fractions. Since the estimated packing fraction for the TRISO fuel compacts is expected to be 30-38% [25], plots with packing fractions of 30%, 35%, and 40% were used. Figure 6-12 through Figure 6-14 show the results plotted as a function of conductivity ratio ($\kappa$), and once again the results show the same conclusion as Figure 6-9 through Figure 6-11. The unit cell models agree best with the full version of Maxwell’s equation and the random model agrees best with the Chiew & Glandt model. Only the random model results are shown in Figure 6-13 because that particular packing fraction was not studied with the unit cell models.
Figure 6-9. Effective thermal conductivity as a function of packing fraction with a matrix conductivity of $k_m = 30 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$.

Figure 6-10. Effective thermal conductivity as a function of packing fraction with a matrix conductivity of $k_m = 40 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$.
Figure 6-11. Effective thermal conductivity as a function of packing fraction with a matrix conductivity of $k_m = 50 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$.

Figure 6-12. Effective conductivity as a function of thermal conductivity ratio ($\kappa$) at a packing fraction of $\varphi = 30\%$. 

$k = 0.0827$
$k_p = 4.1328 \text{ W/m/K}$
$k_m = 50 \text{ W/m/K}$
Figure 6-13. Effective conductivity as a function of thermal conductivity ratio ($\kappa$) at a packing fraction of $\phi=35\%$.

Figure 6-14. Effective conductivity as a function of thermal conductivity ratio ($\kappa$) at a packing fraction of $\phi=40\%$. 
The previous results were all with particles with no heat generation. In reality the real
TRISO fuel compacts will have particles with heat generation. A comparison of the results
between models with and without heat generation in the particles is shown in Figure 6-15 and
Figure 6-16. The ETC results are almost identical for tests run with heat generation vs. no heat
generation in the kernel. This is surprising considering that the temperatures and temperature
gradients between the two models are quite different. Figure 6-17 shows the two temperature
solutions side by side. The solution without particle heat generation has an overall temperature
difference between the top and bottom of the model of 0.32 K while the solution with particle
heat generation has a temperature difference of 25.75 K. That is a considerable temperature
difference, and thus a considerable increase in the driving force of heat flow, i.e., the temperature
gradient, and yet the ETC of each model is the same. This is intuitive since the discrete sources of
heat from the particles would seem to act like heat sources where the external axial heat would
have to flow around because heat is flowing out of the particles, thus restricting the amount of
pathways the heat could flow. The linear nature of the heat equation (Equation (4.1)) shows that
the heat generation term cannot change the properties of the material. Instead the heat generation
just adds to the net heat flux through the sample resulting in a larger temperature gradient. So the
heat generation of the particles does not have an effect on the ETC, but it will have an effect on
the temperature distributions in the TRISO fuel compact.
Figure 6-15. Comparison of unit cell model results for tests with and without heat generation in the kernels.

Figure 6-16. Comparison of random model results for tests with and without heat generation in the kernels.
Figure 6-17. Temperature gradient comparison between particles with no heat generation vs. particles with heat generation, respectively.
For the experimental results the main conclusion is that the matrix material developed by the Germans is not the same as the matrix material being developed for the NGNP program. Therefore it is not suggested to use the German data in the PARFUME calculations. Some of the other conclusions are:

- The thermal conductivity measurement system has been validated and can measure samples with thermal conductivities in the range of 3-70 W·m⁻¹·K⁻¹ with an uncertainty of < 6%. The validation sample measurement deviations are < 6-8% from literature values.

- The matrix samples showed a similar trend as the German data, with a decreasing thermal conductivity from 30-10 W·m⁻¹·K⁻¹ in the temperature range from 100-700°C. The values are lower than the German data, which varies from 45-30 W·m⁻¹·K⁻¹ in the same temperature range. The variation in thermal conductivity between samples is large, around 20-30 W·m⁻¹·K⁻¹ for different matrix types. Even from the same lot of samples the variation can be more than 20%. This is likely because the cracks can cause large thermal conductivity differences in like samples.

- The thermal conductivity of the surrogate fuel samples was found to vary from 40-50 W·m⁻¹·K⁻¹ at 100°C to 25-30 W·m⁻¹·K⁻¹ at 600°C, with a similar decreasing trend as the German data. The higher thermal conductivity of the surrogate samples is unexpected and cannot be explained at this time.
The results from the numerical calculations show that there are better effective conductivity models to use. The model currently being used is the reduced Maxwell equation, but other models show better agreement.

The packing type has very little effect upon the effective thermal conductivity.

A homogeneous thermal conductivity for the TRISO particles can be approximated as 4.1328 W·m⁻¹·K⁻¹.

Heat generation in the particles does not have a significant effect on the effective thermal conductivity of the TRISO fuel compacts.

The full version of Maxwell’s equation agrees very well with the results from the models based upon a SC, BCC, or FCC unit cell.

The Chiew & Glandt model agrees best with the random model results.
CHAPTER 8

FUTURE WORK

So far the effective thermal conductivity of the TRISO fuel compacts has been shown to follow the Maxwell and Chiew & Glandt models. But in order for this conclusion to be validated real samples would need to be tested. This can only be done if the right kinds of samples are sent to Utah State University to be tested. In order to validate the conclusion that the Maxwell and Chiew & Glandt equations correlate well with real TRISO fuel compacts the following samples would be needed:

- A matrix-only sample with the same matrix blend and same resin as the surrogate TRISO fuel compacts
- At least three surrogate TRISO fuel compacts with different packing fractions, preferably ranging from 20-40%

If these samples could be tested then it could be concluded whether or not the TRISO fuel compacts effective thermal conductivity will correlate well with the Maxwell and Chiew & Glandt equations. If the results do not correlate then the data could be used to develop a new effective conductivity model to be used for this specific application.

Future work for the numerical model would be to include temperature dependent thermal conductivities of all the materials. This would improve upon the current COMSOL model to make it match the real world scenarios that exist inside the TRISO fuel compacts. This could also be used to determine a temperature dependent homogeneous thermal conductivity value for the particles. Other factors can also be investigated, including:

- Effect of interfacial resistances between TRISO layers as well as between the particles and the matrix
The researchers at ORNL that make the samples explained that the reason for the higher thermal conductivity of the surrogate samples could result from an enhanced thermal conductivity at the particle surface due to aligning of the graphite particles to create a better heat flow path around the particles. If this phenomenon is real the effect could be modeled using the numerical models developed.
REFERENCES


Appendix A: Random Point Generation Code

(*Units will be in mm*)
(*Specify the dimensions and packing fraction*)
\[ \text{APF} = 0.18; \]
(*Particle radius size*)
\[ r = 0.66; \]
(*side is 1/3 of height*)
\[ s = \frac{1}{3}; \]
\[ \frac{1}{9}; \]
(*number of spheres for the interior, sides, top, and bottom*)
\[ n_{\text{side}} = 10; \]
\[ n_{\text{top}} = 5; \]
\[ n_{\text{int}} = 100; \]
\[ n_{\text{fullsph}} = 125; \]
(*Specify the range for random values*)
\[ x_{\text{minrange}} = r; \]
\[ x_{\text{maxrange}} = s - r; \]
\[ y_{\text{minrange}} = r; \]
\[ y_{\text{maxrange}} = s - r; \]
\[ z_{\text{minrange}} = r; \]
\[ z_{\text{maxrange}} = 1 - r; \]
(*Solve for height of the prism*)
\[ l = \left( \frac{\frac{4}{3} \pi r^3 n_{\text{fullsph}}}{\text{APF}} \right)^{1/3} \]
\[ s \]
(*Create empty list of the points*)
\[ \text{points} = \{(\text{RandomReal}([x_{\text{minrange}}, x_{\text{maxrange}}]), \}
\[ \text{RandomReal}([y_{\text{minrange}}, y_{\text{maxrange}}]), \text{RandomReal}([z_{\text{minrange}}, z_{\text{maxrange}}]))\}; \]
(*begin a counter for the number of random points generated*)
\[ i = 1; \]
\[ k = 0; (*k is an iteration counter*) \]
(*While loop to generate the interior points*)
\[ \text{While}[i < n_{\text{int}}, \]
p = (RandomReal[{xminrange, xmaxrange}],
    RandomReal[{yminrange, ymaxrange}], RandomReal[{zminrange, zmaxrange}]);
k++;

(*dist is where the distances from the
   current point to all the previous points is stored*)
dist = {};
Do[
    AppendTo[dist,
        Sqrt[(p[[1]] - points[[j, 3]])^2 + (p[[2]] - points[[j, 2]])^2 + (p[[3]] - points[[j, 1]])^2]],
        {j, l, i}];
(*If the number of iterations is too large then the interior points operation ends*)
If[k > 100000, Break[]];
(*Checking dist to make sure that the point will be valid,
   and if so then it prints some output to the screen and resets the iteration counter*)
If[Min[dist] > (2*r + .01), AppendTo[points, p]; i++; Print[{{i, k}}]; k = 0];
]
Print[];

(*These next operations do the same as above but at the four sides, top, and bottom*)
k = 0;
While[1 < nint + nsides,
p = {0, RandomReal[{yminrange, ymaxrange}], RandomReal[{zminrange, zmaxrange}]};
k++;
dist = {};
Do[
    AppendTo[dist,
        Sqrt[(p[[1]] - points[[j, 3]])^2 + (p[[2]] - points[[j, 2]])^2 + (p[[3]] - points[[j, 1]])^2]],
        {j, l, i}];
If[k > 50000, Break[]];
If[Min[dist] > (2*r + .01), AppendTo[points, p]; i++; Print[{{i, k}}]; k = 0];
]
Print[];

k = 0;
While[1 < nint + 2 nsides,
p = {z, RandomReal[{yminrange, ymaxrange}], RandomReal[{zminrange, zmaxrange}]};
```mathematica
k++; dist = {}; Do[
    AppendTo[dist,
        Sqrt[(p[[3]] - points[[j, 3]])^2 + (p[[2]] - points[[j, 2]])^2 + (p[[1]] - points[[j, 1]])^2]],
        {j, 1, 4}];
If[k > 50000, Break[]];
If[Min[dist] > (2 x + .01), AppendTo[points, p]; i++; Print[{i, k}]; k = 0];
] Print[];

k = 0;

While[i < nint + 3 nsde,
    p = (RandomReal[{xminrange, xmaxrange}], 0, RandomReal[{zminrange, zmaxrange}]);
    k++; dist = {}; Do[
        AppendTo[dist,
            Sqrt[(p[[3]] - points[[j, 3]])^2 + (p[[2]] - points[[j, 2]])^2 + (p[[1]] - points[[j, 1]])^2]],
            {j, 1, 4}];
    If[k > 50000, Break[]];
    If[Min[dist] > (2 x + .01), AppendTo[points, p]; i++; Print[{i, k}]; k = 0];
] Print[];

k = 0;

While[i < nint + 4 nsde,
    p = (RandomReal[{xminrange, xmaxrange}], 0, RandomReal[{zminrange, zmaxrange}]);
    k++; dist = {}; Do[
        AppendTo[dist,
            Sqrt[(p[[3]] - points[[j, 3]])^2 + (p[[2]] - points[[j, 2]])^2 + (p[[1]] - points[[j, 1]])^2]],
            {j, 1, 4}];
    If[k > 50000, Break[]];
    If[Min[dist] > (2 x + .01), AppendTo[points, p]; i++; Print[{i, k}]; k = 0];
] Print[];
```
Print[]; 
k = 0; 

While[i < n - ntop,  
p = (RandomReal[{xminrange, xmaxrange}], RandomReal[{yminrange, ymaxrange}], 0);  
k++;

dlist = {}
Do[
    AppendTo[dlist,  
      Sqrt[(p[[3]] - points[[j, 3]])^2 + (p[[2]] - points[[j, 2]])^2 + (p[[1]] - points[[j, 1]])^2]],  
      {j, 1, i}]; 
If[k > 50000, Break[]];  
If[Min[dlist] >= (2 r + .01), AppendTo[points, p]; i++; Print[{i, k}]; k = 0];
]

Print[]; 
k = 0; 

While[i < n,  
p = (RandomReal[{xminrange, xmaxrange}], RandomReal[{yminrange, ymaxrange}], 1);  
k++;

dlist = {}
Do[
    AppendTo[dlist,  
      Sqrt[(p[[3]] - points[[j, 3]])^2 + (p[[2]] - points[[j, 2]])^2 + (p[[1]] - points[[j, 1]])^2]],  
      {j, 1, i}]; 
If[k > 50000, Break[]];  
If[Min[dlist] >= (2 r + .01), AppendTo[points, p]; i++; Print[{i, k}]; k = 0];
]

Length[points]
points;  
(*Exports the list of points to a CSV file*)  
Export["C:\Users\Charlie\Documents\Thesis\Effective \nK Consol Model\Random Rectangle\18\rectpoints_18.csv", points]
```plaintext
(* This performs a second check to make sure no particles will overlap *)
ll = Length[points]
For[t = 1, t < ll, t++,
   (* Print[t]; *)
   clist = {};
   Do[
      AppendTo[clist, Sqrt[(points[[t, 3]] - points[[j, 3]])^2 +
                         (points[[t, 2]] - points[[j, 2]])^2 + (points[[t, 1]] - points[[j, 1]])^2]],
      {j, 1, t - 1}];
   Do[
      AppendTo[clist, Sqrt[(points[[t, 3]] - points[[j, 3]])^2 +
                         (points[[t, 2]] - points[[j, 2]])^2 + (points[[t, 1]] - points[[j, 1]])^2]],
      {j, t + 1, ll}]
   If[Min[clist] < 2 r, Print[t]]
]

(* Graphical representation of what the geometry will look like *)
Graphics3D[{Sphere[points, r], Opacity[.3], Cuboid[({0, 0, 0}, {s, s, 1})]}]
```
Appendix B: LiveLink for MATLAB Code

(*Imput the packing fraction and solve for the rectangular prism size*)
APF = 0.23;
r = .46;

\[
\text{height} = \left( \frac{1}{2} \pi r^2 \times 9 \times \frac{1}{\text{APF}} \right)^{1/3}
\]
side = \frac{\text{height}}{3}
12.5872
4.19573

(*Import the list of random point locations*)
xyz = Import["C:\\Users\\Charlie\\Documents\\Thesis\\Effective\
K Consol Model\\Random Rectangle\\24\\rectpoints_24.csv"]; nparticles = Length[xyz];

(*The name and location of the output text file*)
fname = "C:\\Users\\Charlie\\Documents\\Thesis\\Effective\
K Consol Model\\Random Rectangle\\24\\Random_0.24.txt";

(*Generate the LiveLink for MATLAB function to generate the geometry*)
s = OpenWrite[fname]; WriteString[s, "function out-model\n"]; WriteString[s, "import com.comsol.model.x\n"] WriteString[s, "import com.comsol.model.util.x\n"] WriteString[s, "\n"] WriteString[s, "model = ModelUtil.create('Model');\n"] WriteString[s, "\n"] WriteString[s, "\nmodel.modelPath('C:\COMSOL44\models\COMSOL_Multiphysics\Multiphysics');\n"] WriteString[s, "\n"] WriteString[s, "\nmodel.modelNode.create('mod1');\n"] WriteString[s, "\n"] WriteString[s, "\nmodel.geom.create('geom1', 3);\n"] WriteString[s, "\nmodel = ModelUtil.create('Model');\n"] WriteString[s, "\n"] WriteString[s, "\nmodel.name('Random.mph');\n"] WriteString[s, "\n"]

(*Create the Parameters*)
WriteString[s, "model.param.set('Kernel_radius', '0.25[mm]');\n"]
WriteString[s, "model.param.set('radius', '46[mm]');\n"]
WriteString[s, "model.param.set('Matrix', '40[W/m/K]');\n"]
WriteString[s, "model.param.set('Temperature_Low', '373.15[K]');\n"]
WriteString[s, "model.param.set('Temperature_High', '423.15[K]');\n"]
WriteString[s, "model.param.set('NHeatRate', '3[W]');\n"]
WriteString[s, "model.param.set('NHeat_Generation', '3.288[W/m^3]');\n"]
WriteString[s, "model.param.set('NHeat_Generation*Kernel_radius^3/radius^3');\n"]
WriteString[s, "model.param.set('Side', 'S', 'side', '3[mm]');\n"]
WriteString[s, "model.param.set('Height', '+', 'height', '3[mm]');\n"]
WriteString[s, "model.param.set('Area', 'Side^2');\n"]
WriteString[s, "model.param.set('Volume', 'Side^2*Height');\n"]
WriteString[s, "model.param.set('NParticles', '+', 'nparticles', '');\n"]
WriteString[s, "\n"]
WriteString[s, "model.modelNode.create('mod1');\n"]

(*Creating the Geometry*)
WriteString[s, "model.geom.create('geom1', 3);\n"]
WriteString[s, "\n"]
WriteString[s, "model.geom('geom1').lengthUnit('mm');\n"]
WriteString[s, "model.geom('geom1').feature.create('blk1', 'Block');\n"]
WriteString[s, "model.geom('geom1').feature('blk1').setIndex('size', 'Side', 0);\n"]
WriteString[s, "model.geom('geom1').feature('blk1').setIndex('size', 'Side', 1);\n"]
WriteString[s, "model.geom('geom1').feature('blk1').setIndex('size', 'Height', 2);\n"]
WriteString[s, "\n"]

Do[
(*Creating the spheres at the random point locations*)
WriteString[s, "model.geom('geom1').feature.create('sph', j, '+', 'Sphere');\n"]
WriteString[s, "model.geom('geom1').feature('sph', j).set('pos', AccountingForm[xys[[j, 1]], 20, NumberSigns -> "-", ""],\n** "", AccountingForm[xys[[j, 2]], 20, NumberSigns -> "-", ""], ** "",\nAccountingForm[xys[[j, 3]], 20, NumberSigns -> "-", ""], "{")];\n"]
WriteString[s, "\n"]
Do[
(*Creating all the intersection boolean operations*)
WriteString[s, "model.geom('geom1').feature.create('int', j, '+', 'Intersection');\n"]
}
WriteString[s, "model.geom('geom1').feature('int', j, '').set('keep', true);\n"];
WriteString[s, "model.geom('geom1').feature('int', j, '').select('input').set(['blk1', 'sph*', j, '"']);\n"];
WriteString[s, "\n"];
WriteString[s, "l_mparticles\n"]
}
WriteString[s, "\n"]
WriteString[s, "model.geom('geom1').feature.create('diff1', 'Difference');\n"]
WriteString[s, "model.geom('geom1').feature('diff1').selection('input').set(['blk1']);\n"]
WriteString[s, "\n"]
WriteString[s, "out = model;\n"]
Close[s];