THERMAL CONDUCTIVITY MEASUREMENT TECHNIQUE AND APPARATUS DEVELOPMENT
FOR APPLICATION TO TRANSURANIC NUCLEAR FUEL MATERIALS

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Abstract

The advancement of new nuclear fuel compositions requires accurate knowledge of the material’s nuclear and thermal properties. The use of mixed fuel compositions could transmute fission products into lower level waste. However, the thermal properties of new fuel compositions require better understanding. The purpose of the research was to investigate the applicability of an adapted four-point probe (4PP) electrical resistivity measurement technique to determine the thermal conductivity of metallic fuels and to develop a technique and apparatus that could be used to make the measurements within a Hot Cell. In this study, a standard 4PP was used to measure the electrical resistivity of the material and determine the thermal conductivity using the Wiedemann-Franz Law. The 4PP was selected for its resistance to radiation damage and ease with which it could be used in a Hot Cell. The thermal conductivity of Steel and Aluminum samples of various geometries were measured and compared to known values to validate the approach. The approach was found to accurately measure the thermal conductivity within 5 W/m-K. The technique and apparatus developed in this study can be used in Hot Cells to measure the thermal properties of new nuclear fuel compositions.
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Chapter 1: Introduction

Background

In 2017, global energy consumption was 15.7 PWh ($10^{15}$ Watt/hours) of energy, the equivalent of 13.5 billion tons of oil [1], and global carbon dioxide emission was over 36 Giga-tons of CO$_2$ [2]. Meeting this energy need is both resource intensive and produces large quantities of waste. Energy demand is expected to increase 30% by 2040 [3], according to the International Energy Agency. Most of the energy production comes from the combustion of the fossil fuels: coal, natural gas, and oil. To sustainably meet the energy needs of the future a diversified energy portfolio is needed that produces abundant energy, from few resources, with little waste. The energy density of nuclear fuel is so high, and nuclear fuel resources so abundant, that fission is an effectively unlimited energy source for the future.

Figure 1 below depicts the energy profile of the United States in 2017. The United States produces nearly two thirds of its energy, 63%, from fossil, 20% from nuclear, and only 7% from wind and solar combined.

Increased interest in renewable energy has seen an increase in total energy production in recent years, but still contributes only a small portion of total production. Nuclear energy contributes over half of all
clean energy. For a number of reasons, renewable’s alone will be inadequate to meet the energy demands of the future and a significant increase in nuclear infrastructure is needed in the United States and globally.

But nuclear infrastructure in the United States is aging with the average reactor age of 37 years. Most of the reactors currently in operation in the U.S. are Generation II. Modern reactor technology is characterized as generation III+ and represents significant evolutionary improvements on the original generation II concepts.

### Evolution of Nuclear Power

Current focus is now moving toward revolutionary generation IV nuclear reactor concepts. Generation IV is significantly different from current technology and seeks to improve efficiency, safety, proliferation resistance, cost, and waste mitigation.

Fresh nuclear fuel used in Generation II reactors is composed of UO$_2$, where the Uranium is approximately 3-5% fissile U$^{235}$ and 97-95% fertile U$^{238}$. After about 2-3 years spent fuel is removed from the reactor core and is composed of fission products, leftover U$^{235}$ that did not undergo fission, Plutonium transmuted from the fertile U$^{238}$, and the remaining U$^{238}$. The Plutonium, Pu$^{239}$, is itself fissile and contributes a significant portion of the fission energy towards the end of the fuels time in the core.
In an open fuel cycle the spent fuel is then stored as nuclear waste. However, the $\text{U}^{235}$ and $\text{Pu}^{239}$ can be used as fuel and reprocessed along with the $\text{U}^{238}$. Recycling nuclear fuel is a proven concept that is practiced outside the US. The only waste in a closed fuel cycle are the fission products. The radiological hazard of a radioactive material is related to its half-life and the radiation it gives off. High level waste must be stored for very long periods, on the order of a thousand years, were as low level waste only needs to be stored for decades. Therefore, there is interest in transmuting fission products from high level waste to low level waste. In principle the fission product can be mixed into the recycled fuel and burned to reduce the amount of nuclear waste. The composition of such a fuel is an active area of research so generation IV reactors could be designed to run on the fuel. However, nuclear reactor design requires accurate knowledge of the material properties of the nuclear fuel, in particular the thermal conductivity. There is a need to be able to measure the thermal conductivity of these new mixed fuel compositions.

Nuclear materials, however, are radioactive which creates additional difficulties measuring material properties. To shield experimenters from the radiation, nuclear materials are handled in a containment unit known as a hot cell. The operator uses mechanical arms to remotely handle the materials. The robotic manipulators are difficult to operate and limit what experiments can be
implemented in that environment. An additional difficulty testing radioactive materials is the apparatus must not be sensitive to radiation damage. Thus, the method used to measure the thermal conductivity of new nuclear fuel compositions needs to be easy to conduct using remote handling and must not be sensitive to radiation damage.

**Thermal Conductivity Measurement Techniques**

A variety of experimental approaches exist to measure thermal conductivity but are either difficult to conduct remotely and or require sensitive equipment. For thermal characterization of bulk material, five primary methods are employed [7]:

1) The steady-state method
2) Transient hot-wire method
3) Laser flash diffusivity method
4) Transient plane source method
5) \(3\omega\)-method

and

6) Thermoreflectance methods

In the steady state method the sample placed between two plates of known conductivity and brought up to a constant temperature before measuring the thermal conductivity. The limitations of this approach are the difficulty in setting up the apparatus, that the sample must be heated, that the sample is assumed to have uniform conductivity, and that the method is slow.

In transient hot-wire method a wire imbedded in the material acts as a heat source to uniformly heat the material. [8] Temperature measurements are then taken on the outside of the material to determine the temperature change with respect to time, thereby determining how quickly heat conducted within the material. A limitation in this approach is that the heating wire must be imbedded
in the material, which would present fabrication difficulties for the proposed conductivity measurements. Additionally, the material must be heated.

The laser flash diffusivity method uses a concentrated photon beam to impart energy to one side of a sample while the temperature is measured on the other side of the sample to determine thermal diffusivity. This approach could be used for the proposed conductivity measurements, but consideration would be needed to insure the equipment was shielded from radiation. Positioning the sample correctly using remote handling may also be difficult, as would obtaining samples large enough to use this method [9].

In the transient plane source method a sensor, made of conductive material of known thermal and electrical properties, is placed between two halves of a sample and current is applied. As the sensor heats, heat is conducted away by the sample. By measuring the temperature of the sample over time the rate at which the sample conducted heat can be determined [10]. Primary limitation of this approach is the required sample size, which may be large for materials with high conductivities. Additionally, the sample must be cut into two pieces to place the sensor between. Which presents fabrication difficulties. An inability to make localized conductivity measurements is another limitation.

Finally, in the 3ω-method a material with a known thermo-electric constant is applied to the surface of the sample to act as both a resistive heater and a Resistance Temperature Detector (RTD). An AC current is applied to the wire at a certain frequency causing joule heating. Heat conduction within the material causes a subsequent temperature change in the sample and RTD. Through careful measurement of voltage in the RTD and analysis the phase lag, corresponding to the rate of conduction, can be determined. This technique present advantages over the other methods and has been found to return good results, but presents difficulties in fabrication, because the RTD must be deposited on the sample, and in difficulties arising from electrical conduction within the sample.
Proposed Approach

Overall many different methods exist to measure thermal properties of materials. Almost all the methods of measuring thermal conductivity involve heating the sample and measuring the thermal conduction. Heating of the sample is not desirable because it is harder to implement and is not NDT. Many of the methods present fabrication difficulties and or additional consideration to shield the equipment.

The proposed approach, in an attempt to mitigate the limitations of existing methods, is to use an electrical resistivity measurement technique to determine the electrical conductivity of the material. The electrical and thermal conductivity of a material are fundamentally related through the Wiedemann-Franz law. Using electrical contact to measure resistivity is easier than measuring thermal conductivity, will not require sensitive equipment or difficult sample preparation, and is more position insensitive than the thermal conductivity measurement methods.
Fundamentally, the movement of free valence electrons within metals causes thermal transport. Movement of free electrons in a metallic material conducts electrical energy in the presence of a voltage potential. Conversely, movement of free electrons in a metallic material conducts thermal energy in the presence of a temperature potential. An increase in temperature increases lattice collisions, increasing electrical resistivity and decreasing electrical conductivity $\sigma$. An increase in temperature increases lattice collisions, decreasing thermal resistivity and increasing thermal conductivity $\kappa$. Therefore, Electrical conductivity is inversely proportional to temperature and thermal conductivity is directly proportional to temperature. Gustav Wiedemann and Rudolph Franz observed that both are thus proportional to temperature. Further, the ratio of the conductivities is proportional to temperature, regardless of material. Experiments revealed an empirically determined proportionality constant, known as the Lorenz number $L$, to relate the ratio of the conductivities to temperature. Thus, the basic Wiedemann-Franz Law was derived.

$$\frac{\kappa}{\sigma} \propto LT$$

Later developments in quantum mechanical models allowed for the derivation of the theoretical value for the Lorenz number.

Band Theory is fundamental to the quantum mechanics and states that electron can occupy only specific energy bands around an atom, forming valance bands. When the atoms are bound in a solid lattice a conductive band is formed between atoms. This low energy band allows electrons to flow freely between the atoms. The conductive bands dictate the electrical and thermal properties of materials. The Drude-Lorentz model is a model based on kinetic theory that explains electrical conduction in terms of movement and collision of electrons in the material. The model considers the random motion and collisions of electrons within a material and the applied electric field to determine the net movement of charge through the material [11].
\[ J = \left( \frac{nq^2 \tau}{m} \right) E \]  

(2)

Where \( J \) is the current density, \( n \) is the particle density, \( q \) is the electric charge of an electron, \( \tau \) mean collision time, \( m \) is the mass of an electron.

Later, the Drude-Sommerfield model was developed that modeled the free electrons as a gas and, notably, neglected ion-electron collisions. The Free Electron Model, as it is also known, explains much of the observed behavior of electrons and provides a quantum mechanical basis for thermal properties. Taking the heat transfer to be [11]

\[ Q = \frac{mnv dT}{3\lambda c_v} \frac{dT}{dx} \]

(3)

where \( Q \) is the heat flow, \( c_v \) is the specific heat, \( n \) is the number of particles per unit volume, \( \lambda \) is the mean free path and \( dT/dx \) is the temperature gradient. From the free electron model, the energy of free electrons is [11]

\[ mc_vT = \frac{3}{2} k_B T \]

(4)

where \( T \) is temperature in Kelvin and \( k_B \) is the Boltzmann constant. Modeling the electrons as an ideal gas, thermal conductivity \( K \) is defined as

\[ K = \frac{1}{3} c_v \lambda v \]

(5)

And using the above equation for current density from the Drude Model, Sommerfield derived [11]

\[ \frac{K}{\sigma} = \frac{3K_B^2 T}{2e^2} \]

(6)

\[ \frac{K}{\sigma T} = 2.44 \times 10^{-8} \ \Omega W/K^2 = \text{Lorenz number (L)} \]

Where \( e \) is the charge of an electron = 1.502 x 10^{-9} C.

Which is known as the Sommerfield value of Lorentz number and is widely accepted as most accurate theoretical value for the Lorentz number. The Wiedemann-Franz Law is the best modeling comparing electrical and thermal conductivity but has several limitations. First, the Lorenz number is not
independent of material. The Sommerfield value is very close to the experimentally determined Lorentz number of materials and a study investigating the accuracy of the Sommerfield value reported 1.5% experimental error in Lorentz number [12]. The scattering of electrons was cited as the cause of deviations in the Lorenz number and also reported that electron-electron interaction was not found to be significant. A comprehensive review of experiments investigating the Lorentz number complied the findings into tables and showed that the Lorentz number varies between materials, but is relatively constant for conductive metals [13].

A study in 2015 investigated the relation between the Seebeck coefficient, $S$, and the Lorentz number [14]. This allows the Lorentz number, which is material dependent, to be related to a measurable material property, the Seebeck coefficient, which is a measure of the thermoelectric effect in a material. As noted early, this publication noted that the effects of electron scattering can significantly affect the Lorenz number. The relation to the Seebeck coefficient was found to be

$$L = 1.5 + \exp \left[ -\frac{1.05}{1.10} \right]$$

And was reported to be accurate within 5% in semi-conductors. A major limitation to this approach is that the Seebeck co-efficient must first be determined [15], which, for the purposes of measuring the thermal conductivity of new nuclear fuel materials is problematic. An “Apparatus for the high temperature measurement of the Seebeck coefficient in thermoelectric materials” [16] was developed, but the measurement of Seebeck co-efficient requires a complex experimental set-up that would not be applicable to use in a hot cell. Further, the apparatus is susceptible to radiation damage and is more complex then would be needed to measure thermal conductivity directly. Thus, the Sommerfield value will be used for the purposes of this research because it is the most accurate value if nothing else about the material is known.

Finally, perhaps the most significant limitation to the Weidermann-Franz law is that it does not account for Phonon conduction. As stated previously, both electrical and thermal energy is transferred
through metals through the movement of electrons. However, vibrations in the materials lattice also transfers thermal energy. These vibrations propagate similarly to waves and are known as Phonons. The total thermal conductivity is the sum of the conduction due to electrons and the conduction due to phonons. Thus, without knowledge of the phononic conduction the Weidemann-Franz Law cannot be used to determine the total thermal conductivity exactly. However, it is worth noting that phononic conduction is significantly lower than electron conduction such that the thermal conductivity of ceramics is dominated by phonons and the thermal conductivity of metals is dominated by electrons. A study found that Lorenz number deviates from the Sommerfield value at intermediate temperatures due to phonon thermal conductivity [17]. Since the experimental technique under investigation is concerned with metallic fuels the phononic contribution to the thermal conductivity can be considered negligible.

**Electrical Conductivity Measurement Techniques**

A variety of electrical resistivity measurement techniques exist but are all based on the same fundamental principle: Ohm’s Law.

\[ V = IR \]  

By applying a known electric current to the sample and measuring the subsequent voltage drop the resistance of the sample can be measured. Consideration of the sample geometry allows the resistance to be used to determine resistivity.

1. **Two probe**

   One probe is placed on either side of a rectangular block sample. The probes are used to both supply the excitation current and measure the resulting voltage drop.

2. **Four probe**
Two probes are placed on each side of a rectangular block sample. One set of probes supplies the excitation current and the other set of probes measures the resulting voltage drop. This makes the voltage measurement slightly more accurate than two probe.

3. **Four-point probe (linear configuration)**

Four electrical contacts are arranged in a line with uniform pin separation and placed on the surface of the sample. Current is applied to the outside probes causing a voltage gradient to be produced in the sample. The voltage drop across the inside probes is used to measure this voltage gradient. Four-point probe is more accurate than four probe and accommodates a greater variety of sample geometries, provided the sample is rectangular or circular and of uniform thickness. To account for sample geometry Resistivity Correction Factors (RCF) are used.

4. **Four-point probe (rectangular configuration)**

The four probes are placed in a rectangular or square configuration as opposed to a linear configuration. Resistance measurements can then be taken with any combination of the four probes either applying current or measuring voltage. The rectangular configuration is less commonly used and is generally no more accurate than a linear configuration. Calculation of RCF’s presents additional difficulties.

4. **Four point**

The four probes are placed on the circumference of the material allowing for samples of any geometry to be measured. Several conditions must be met to use this approach, such as the sample must be a thin sheet, and is in general more difficult to implement than the four-point probe.

6. **Pulse probe**

Voltage pulses used to measure the voltage or current across the sample. Pulsing is used to reduce the effects of heating. For materials of high conductivity the excitation current does not need to be large, so the effects of joule heating can be considered negligible.
The four-point probe (linear configuration) is by far the most common and is used widely in the semiconductor industry for quality control and testing of semi-conductive material. Several considerations can be taken to improve accuracy of measurements [18]:

1) Scratching or cutting surface to expose fresh material may be necessary if the material is prone to oxidation
2) Cleaning the surface with a good solvent to remove debris
3) Insuring good pin contact by clamping probe to the sample
4) Soldering the probe to the sample
5) Plating of probe and or sample with highly conductivity material to mitigate resistive losses at the interface
6) Calibration of the probe and method(s)
7) Avoiding high current to prevent sample heating
8) Materials exhibit non-linear adherence to Ohm's law for high voltages
9) Reversing the direction of the measurement can be used to verify good pin contact
10) In rare cases light pollution can cause the resistivity of some materials to change

A research study with similar objectives, “A thermal conductivity and electromotive force measurement system for nuclear fuels and materials” [19] used thermocouples and an applied heat source to measure relative changes in thermal conductivity and measure electromotive force. The measurement system also predicts phase transitions in materials. The publication also suggested the approach would be applicable to use in a hot cell. The approach is similar to the thermal conductivity measurement methods discussed previously and has similar limitations. Given existing thermal conductivity and electrical resistivity measurement techniques, an evaluation of each methods limitations, accuracy, and sensitivity and consideration difficulty of remote handling, the four-point probe was selected as the method this research study would investigate.
Four-Point Probe – Theory

Fundamentally, the current density is related to resistivity and voltage potential by [20]

$$\nabla j = \nabla \frac{E}{\rho} = \nabla^2 V = 0. \tag{9}$$

Considering the current field as a semicircle and introducing imaginary poles to account for material boundaries, the voltage at a point \( r \) from the current source can be expressed as [21]

$$V = \frac{lr}{2\pi r^2}. \tag{10}$$

Resistance is proportional to the resistivity, length, and cross section of the material

$$R = \rho \frac{L}{A}. \tag{11}$$

For uniform probe spacing \( s \), the measured resistivity \( \rho_o \) is calculated by [21]:

$$\rho_o = \frac{V}{I} \times 2\pi S. \tag{12}$$

But a RCF, \( f \), is needed to account for boundary conditions [20]

$$\rho = \frac{\rho_o}{f(S)}. \tag{13}$$

The RCF can be found tabulated or can be calculated for common geometries.

Considering proximity to edge boundary conditions the voltage at the two sensing probes will be a factor of the current from the two real probes and the two imaginary probes. Taking \( s \) to be the electrode distance and \( x \) to be the distance to the edge the resistivity can be related to the measured voltage and current by [21]

$$I_{measured} \frac{V_{measured}}{\rho} = 2\pi \frac{1}{s} \left[ \frac{1}{s} - \frac{2}{\sqrt{(2s)^2 + (2x)^2}} + \frac{2}{\sqrt{s^2 + (2x)^2}} \right]^{-1}. \tag{14}$$

This approach however does not account for the thickness of the material.

Considering thickness and edge boundary conditions, for infinite volume \( d > 5s \) and \( t >> s \), the resistivity of a sample is measured as:

$$\rho_o = 2 \pi s \ (V/l). \tag{15}$$
In general, RCF are needed to account for boundary conditions [20]:

\[
\rho = \frac{\pi}{\ln(2)} \left( \frac{V}{I} \right) f_1 f_2.
\]

For an insulating bottom boundary \( f_1 = f_{11} \) given by [20],

\[
f_{11} = \frac{\ln(2)}{\ln \left( \frac{\sinh(t / s)}{\sinh(t / 2s)} \right)}.
\]

\( f_2 \) is given graphically, but for \( d/s >> 1 \) is approximately equal to 1.

Considering other probe placements and boundary conditions. A research study from the 1950’s investigated the RCF for a variety of boundary condition [21]. The findings are widely cited in nearly every publication on RCF for four-point probe because of the RCF’s accuracy.

Perpendicular to non-conducting boundary:

\[
F_{L_{\perp}}(\frac{1}{s}) = \frac{1}{1 + \frac{2}{s} + \frac{1}{s} + \frac{1}{s} + \frac{1}{s} + \frac{1}{s}}.
\]

Parallel to non-conducting boundary:

\[
F_{L_{\parallel}}(\frac{1}{s}) = \frac{1}{1 + \frac{2}{s} + \frac{1}{s} + \frac{1}{s} + \frac{1}{s} + \frac{1}{s}}.
\]

Perpendicular to conducting boundary:

\[
F_{L_{\perp}}(\frac{1}{s}) = \frac{1}{1 + \frac{2}{s} + \frac{1}{s} + \frac{1}{s} + \frac{1}{s} + \frac{1}{s}}.
\]

Parallel to conducting boundary:

\[
F_{L_{\parallel}}(\frac{1}{s}) = \frac{1}{1 + \frac{2}{s} + \frac{1}{s} + \frac{1}{s} + \frac{1}{s} + \frac{1}{s}}.
\]

Thin slice conducting boundary:
Thin slice non-conducting boundary:

\[
G_n \left( \frac{w}{2} \right) = 1 + 4 \sum_{i=1}^{w} (-1)^i \left[ \frac{1}{\sqrt{\left( \frac{1}{w} \right)^2 + (2n)^2}} - \frac{1}{\sqrt{\left( \frac{1}{w} \right)^2 + (2n)^2}} \right].
\]  

(22)

RCF F_2, F_3, and G_7 will be used for this research study and a sensitivity analysis of the effect of inexact probe placement can be found in Chapter 2.

Circumferential probe placement on thin materials. If the four point method is used, the probes are placed along the boundary of the material, the material thickness (d) is known, and the resistance between two sets of probes is measured using four terminal sensing the equation [22]

\[
\exp \left( -\frac{\pi d}{\rho} R_{MN,OP} \right) + \exp \left( -\frac{\pi d}{\rho} R_{NO,PM} \right) = 1,
\]

(24)

Can be solved for \( \rho \) as the only unknown resulting in

\[
\rho = \frac{\pi d}{\ln 2} \frac{R_{MN,OP} + R_{NO,PM}}{2} f, \quad \ldots\]

(25)

where \( f \) is a ratio of \( R_{MN,OP}/R_{NO,PM} \).

Circumferential probe placement presents the advantage of accounting for complex sample geometries but is more difficult to implement in a hot cell because each probe must be carefully placed independently along the edge of the material. Because of this the above RCF’s will be used with a linear four-point probe.

Eddy Current Measurement Technique

A final electrical conductivity measurement technique was considered which uses the principle of eddy currents. According to Faraday’s Law, electrons will move in a material in the presence of a varying magnetic field to oppose the magnetic field.
AC through a coil induces a magnetic field which induces an opposing current in the sample. This causes a measurable change in the impedance of the coil which can be used to calculate the conductivity in the material [23]. The general equation for the eddy current probe takes into consideration the varying magnetic field or relative motion, power dissipation due to eddy currents, and the magnetic permeability and electrical resistivity of the material [24]. The induced voltage, also known as the back emf is

$$\nabla^2 \mathbf{H} = \mu_0 \sigma \left( \frac{\partial \mathbf{M}}{\partial t} + \frac{\partial \mathbf{H}}{\partial t} \right)$$  \hspace{1cm} (27)$$

Mutual inductance in the material generates this back emf in response to the time varying current, creating an impedance in the coil. A phase lag is observed and is due to the conductivity and permeability of the sample. By measuring and analyzing the time voltage the phase lag can be determined and used to find the conductivity of the sample.

This method is well suited for use with non-magnetic metals since the conductivity and permeability cannot be separated unless at frequencies much lower than 40 Hz, which is the lower limit
of a typical impedance analyzer [25]. Depth measurements are also possible with the use of an eddy
current probe but in general either the thickness or the conductivity must be known to determine the
other. Eddy current probes are used in industry for large variety of applications such as to detect
material flaws, sort materials, and to determine heat treatment and surface coats, but is not primarily
used for raw conductivity measurements. Pulsed eddy current testing can be used to measure different
depths in the material with uncertainties as low as 0.5% reported [26].

Factors that affect results of eddy current testing include, material conductivity, permeability,
frequency of the AC, geometry of the sample, proximity to edges and lift-off [27]. One limitation is that
conductivity and permeability are difficult to separate, so the approach has limited application to
magnetic materials [28] and the materials permeability must be assumed to calculate the conductivity.
Another limitation is that the samples must be much larger then probe, because the magnetic field is
larger than probe. The approach also requires three data points per measurement for calibration and
must be compared to a reference sample of known conductivity [29]. The approach is sensitivity to
temperature and edges effects.

A research study most applicable for the proposed conductivity measurement invested the
impedance of a coil on the outside of a cylinder. The technique returned good results for infinite length
and radii of curvature greater than 20mm but exhibited significant deviation at 5mm radius. The
publication stated the limitation to this approach which summarizes why eddy current probes were not
selected for this research study. "An inherent drawback of thin coils is their low inductance which means
that the coils often need many windings resulting in a larger footprint and decreased position resolution
for defect detection than conventional coils. The lower inductance also limits the lower frequency
application of such sensors for the detection of buried defects. However, for many applications this is not
important." [30]. Thermal conductivity measurement eddy current probes are available commercially
but require large samples and are not intended for precise thermal conductivity measurements.
Specific Goals of This Research

The specific outcome goals of this research are:

- Insuring good pin contact on material substrate
- Accurate Resistivity Correction Factors to determine electrical resistivity
- Accurate model to relate conductivities based on the Wiedemann Franz Law
- Measurement tool and apparatus applicable to use in hot cell

A commercially available four-point probe was selected which has spring loaded pins to insure good pin contact. Chapter 2 presents a sensitivity analysis of the Resistivity Correction Factors, Chapter 3 presents preliminary results investigating the validity of the approach using an applied DC current. Chapter 4 presents the results investigating the use of a lock-in amplifier with an applied AC current. Chapter 5 concludes with a summary of the results, an evaluation of the applicability of this approach, and recommendations for future work.

Chapter 2: Sensitivity Analysis of Resistivity Correction Factors

In order to assess the validity of the 4PP approach an analysis of the RCF was conducted. Ideally, the RCF would be insensitive to inexact probe placement on the sample as remote handling within a Hot Cell makes exact placement more difficult. Four geometric parameters influence the measured resistivity of the sample: probe spacing, sample thickness, perpendicular distance from edge, and parallel distance from edge.
Figure 5 shows the RCF’s stated above for non-conductive boundaries. As expected, the RCF approaches 1 as the distance from the boundary goes to infinity.

To assess error in RCF due to error in position the ratio of the RCF for probe placement in the exact center of the sample and the RCF for the probe off center was calculated as the departure from the exact center varied from one edge to the other and is plotted in Figure 7 below for the perpendicular RCF.

*Figure 6: Plot of Resistivity Correction Factors [Valdes, #] for Probe Spacing of 1.27mm*
Figure 7: Perpendicular RCF Sensitivity for Probe Spacing $s$ and Sample Width 5mm, One Boundary

Significant error is observed for large departure from true position, as is expected, with the RCF too low if the probe is placed too near the edge and too large if the probe is placed further from the edge. Placement of the probe near the edge of the sample results in a higher measured voltage and thus a higher calculated resistivity. The RCF is less than 1 and becomes smaller as the probe approaches the sample boundary, correcting for the higher voltage. If the probe is placed closer to the boundary than expected the RCF will be too large resulting in an erroneously large resistivity. Conversely, if the probe is placed further the boundary than expected the RCF will be too small resulting in an erroneously small resistivity. Expressed as a percentage, the resistivity is actually some percent of the measured resistivity. This is observed in the sensitivity plot where the actual resistivity is lower than the measured resistivity closer the edge and higher than the measured resistivity further from the edge. However, since the sample is of finite size the RCF should be considered for both ends of the sample.
Figure 8: Perpendicular RCF Sensitivity for Probe Spacing $S$ and Sample Width 5mm, Two Boundaries

The result is an overall reduction in maximum error, a decreased sensitivity, and a measured resistivity that is equal to or greater than the true value. However, sensitivity decreases as the sample size increases as can be seen in Figure 9 below.

Figure 9: Perpendicular RCF Sensitivity

No RCF is associated with probe spacing, however, it is subject to error arising from the manufacturing tolerance of the probe. Sensitivity to inexact probe spacing was calculated for the perpendicular RCF and is plotted in Figure 10. The probe selected for study had a pin separation of
1.27mm and a positional tolerance of +/- 1% for the pins. Probe spacing sensitivity and positional tolerance are shown in Figure 11.

Error from the positional tolerance of the probe was found to be 0.11%. The sensitivity of the parallel and thickness RCF’s are shown in Figure 12-Figure 15.
Figure 12: Parallel RCF Sensitivity for Probe Spacing Of 1.27mm, 5mm Sample Size

Figure 13: Parallel RCF Sensitivity for Probe Spacing Of 1.27mm, Varying Sample Size
Figure 14: Thickness RCF Sensitivity for Probe Spacing Of 1.27mm, Thickness 1.27mm

Figure 15: Thickness RCF Sensitivity for Probe Spacing Of 1.27mm, Varying Thickness

Note that the thickness RCF considers inexact measurement in the thickness of the sample and is therefore only one sided. Relative departure from measured thickness is also expected to minimal.
Figure 16: Comparison of sensitivity to departure from true position for the geometric factors
Figure 16 shows a comparison of the relative sensitivity. Inexact probe spacing and thickness measurement were found to be the most sensitive, but fortunately are also more controllable. The 4PP technique is most sensitive to probe placement parallel to the sample edge.

In practice, large samples have low relative departure from true position resulting in low sensitivity and small samples have large RCF’s resulting in low sensitivity to absolute departure.
Chapter 3: Experimental Results – DC Measurement

Experimental Apparatus and Setup

Following the theoretical analysis of the RCF the four-point probe was tested experimentally on steel and aluminum samples of varying geometries to investigate the sensitivity of the four-point probe to various conditions that might affect the measured conductivity.

The Signatone Four-Point Hand Held probe was used for this study with 1.27mm probe spacing. The probe was mounted on the end of a cantilevered beam with the fulcrum restricted to one degrees of freedom; rotation about an axis normal to the beam and tangential to the probe. This mitigates positional inaccurately and the difficulty of probe placement in remote handling applications. A puck mounting system was employed to hold the samples on pucks of 15mm diameter allowing samples to be easily interchanged while maintaining probe placement in the center of the puck. Figure 18 depicts a computer model rendering of the experimental setup. The weights allow the probe pressure to be varied, although this functionality was not used in this study as discussed later.

![Figure 18: Computer Rendering of Experimental Apparatus](image)

The following effects were considered. The measured resistive was not expected to depend on the applied current because the resistivity is calculated from the measured voltage divided by the applied current, which should scale linearly. The sample geometry was expected to affect the measured resistivity, however, the RCF were expected to account for this. Electrical noise in the system increases measurement uncertainty and it was expected noise might distort the micro scale voltage. The
repeatability of the conductivity measurement was also tested. The four electrical contact pins in the pins are symmetrical so the polarity of the applied DC current so the measured conductivity was not expected to depend on probe polarity. The measured conductivity as also not expected to depend on the overall sample volume, again because the differences in measured resistivity are corrected by the RCF. The probe was also tested on an Aluminum sample and the results compared to the steel sample.

Applied Current
A rectangular steel sample of dimension 25x25x44mm was measured with three different orientations to consider three different RCF conditions. The probe was placed parallel to the 45mm dimension, perpendicular, and normal as depicted in Figure 19 below.

![Figure 19: Illustration of Probe Placement Configurations Relative to Rectangular Sample](image)

The resistivity was measured for each configuration with the applied current varied from 5mA up to 1A. The results are plotted in Figure 20 on the next page. As a comparison the resistivity of the sample was also measured using the two-point probe method where the current and voltage are simulations measured from the same pin. The resistivity was measured across both the 25x25 and 25x45 cross sections of the sample.
Figure 20: Measured Resistivity Response to Applied Current

Table 1: Experimental results of applied DC current

<table>
<thead>
<tr>
<th>Probe Configuration</th>
<th>Mean Resistivity [Ohm-m 10^-6]</th>
<th>Standard Deviation</th>
<th>Thermal Conductivity [W/m-K]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Parallel</td>
<td>0.2127</td>
<td>0.0154</td>
<td>27.4</td>
</tr>
<tr>
<td>Perpendicular</td>
<td>0.1857</td>
<td>0.0056</td>
<td>36.7</td>
</tr>
<tr>
<td>Thickness</td>
<td>0.2043</td>
<td>0.0159</td>
<td>33.3</td>
</tr>
<tr>
<td>Small x-section</td>
<td>0.1563</td>
<td>0.0154</td>
<td>-</td>
</tr>
<tr>
<td>Large x-section</td>
<td>0.1504</td>
<td>0.0125</td>
<td>-</td>
</tr>
</tbody>
</table>

As can be seen from the graph the uncorrected resistivity measured for each configuration was found to vary but remained relatively constant above 0.2A within the expected range of resistivity for steel.

Sample Geometry

The above data is plotted in Figure 21 to show the voltage-current relationship, which is expected to be linear.
The linear relationship was found to hold. Taking the slope of the above graph for each configuration as V/I, calculating the resistivity and applying the RCF’s in Table 1, the configuration averaged resistivity is superimposed on Figure 22 below.

Table 2: RCF for Sample Geometry Test

<table>
<thead>
<tr>
<th></th>
<th>Parallel</th>
<th>Perpendicular</th>
<th>Thickness</th>
</tr>
</thead>
<tbody>
<tr>
<td>RCF – Parallel</td>
<td>0.9989</td>
<td>0.9990</td>
<td>0.9990</td>
</tr>
<tr>
<td>RCF – Perpendicular</td>
<td>0.9992</td>
<td><strong>0.9999</strong></td>
<td>0.9992</td>
</tr>
<tr>
<td>RCF – Thickness</td>
<td>0.999</td>
<td>0.999</td>
<td><strong>1.000</strong></td>
</tr>
<tr>
<td>RCF - Total</td>
<td>0.9989</td>
<td>0.9987</td>
<td>0.9982</td>
</tr>
</tbody>
</table>

Figure 21: Voltage Current Relationship for Sample Geometry Test

Figure 22: Resistivity Dependence on Sample Geometry
As can be seen in Figure 22 the application of the RCF’s did not remove the sample geometry dependence of the measured resistivity. As can be seen by the values in Table 2 the RCF are nearly unity because the sample is large relative to the probe spacing. Figure 23 shows the calculated thermal conductivity for the measured resistivity in Figure 22 and Table 3 tabulates the results.

![Measured Thermal Conductivity Dependence on Applied Current](image)

**Figure 23: Thermal Conductivity Dependence on Sample Geometry**

<table>
<thead>
<tr>
<th></th>
<th>Resistivity [µΩ-m]</th>
<th>Conductivity [W/m-K]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Parallel</td>
<td>27.4x10^{-6}</td>
<td>27.4</td>
</tr>
<tr>
<td>Perpendicular</td>
<td>19.7x10^{-6}</td>
<td>36.7</td>
</tr>
<tr>
<td>Thickness</td>
<td>21.2x10^{-6}</td>
<td>33.3</td>
</tr>
</tbody>
</table>

**Table 3: Resistivity and Thermal Conductivity for Geometry Test**

**Electrical Noise**
An evident concern from the applied current and sample geometry testing is increased variability in the measured voltage at low currents. To investigate the repeatability of measurements the 9mm³ sample was measured on each side at 10mA at a 100 Hz sampling frequency. Voltage measurements were also taken at 0mA to better characterize the electrical noise in the system. Figure 24 shows the voltage measurements on one side for illustrative purposes, the other sides had similar...
results. An offset voltage was observed for the 0mA measurements and the measurements at both 0mA and 10mA varied by 0.5-1 µV.

![Graph showing voltage measurements for repeatability and noise characterization at 10mA.](image)

*Figure 24: Example of Voltage Measurements for Repeatability and Noise Characterization 10mA*

Because the noise in the system was greater than the voltage signal the measured conductivity varied significantly from side to side and returned unlikely thermal conductivity measurements. When using a four-point probe it is generally advised to use a low current. 1-10mA current is typically used for resistivity measurements of semi-conductive material. However, because the metal sample being tested has a significantly lower resistivity the measured voltage drop is smaller, and the effect of noise is more significant. The results of the experiment show that conductivity measurement is inconsistent at 10mA.

<table>
<thead>
<tr>
<th>Side</th>
<th>Measured Voltage [µV]</th>
<th>Conductivity [W/m-K]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Side 1</td>
<td>0.7505 (σ = 0.52)</td>
<td>12</td>
</tr>
<tr>
<td>Side 2</td>
<td>0.3974 (σ = 0.66)</td>
<td>23</td>
</tr>
<tr>
<td>Side 3</td>
<td>0.4281 (σ = 0.75)</td>
<td>21</td>
</tr>
<tr>
<td>Side 4</td>
<td>0.1675 (σ = 0.53)</td>
<td>54</td>
</tr>
<tr>
<td>Side 5</td>
<td>0.0008 (σ = 0.50)</td>
<td>1082</td>
</tr>
</tbody>
</table>

*Table 4: Results of Repeatability Testing and Noise Characterization at 10mA*
Repeatability

To mitigate the impact of noise the applied current was increased to 80mA to increase the measured voltage drop one order of magnitude above the noise while still applying minimal current. Figure 25 shows the voltage measurements on one side for illustrative purposes, the other sides had similar results. Table 5 tabulates the measured conductivity for each side.

![Voltage Measurements](image.png)

**Table 5: Results of Repeatability Testing at 80mA**

<table>
<thead>
<tr>
<th>Side</th>
<th>Measured Voltage [µV]</th>
<th>Conductivity [W/m-K]</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>(σ = standard deviation)</td>
<td></td>
</tr>
<tr>
<td>Side 1</td>
<td>1.844 (σ = 0.58)</td>
<td>39.1</td>
</tr>
<tr>
<td>Side 2</td>
<td>2.681 (σ = 0.53)</td>
<td>26.9</td>
</tr>
<tr>
<td>Side 3</td>
<td>2.639 (σ = 0.53)</td>
<td>27.3</td>
</tr>
<tr>
<td>Side 4</td>
<td>2.100 (σ = 0.65)</td>
<td>34.4</td>
</tr>
<tr>
<td>Side 5</td>
<td>2.683 (σ = 0.50)</td>
<td>26.9</td>
</tr>
<tr>
<td>Side 6</td>
<td>2.491 (σ = 0.50)</td>
<td>29.0</td>
</tr>
</tbody>
</table>

As can be seen from the figure, the variation in the measured voltage was comparable to the results for 10mA but the variation in the calculated conductivity was significantly reduced. The probe was rated
for up to 100mA and increasing the applied current to 80mA was found to significantly increase the repeatability of the conductivity measurement.

**Probe Polarity**

To test the effect of probe polarity one face of the 10mm³ was measured. The polarity of the DC current was then reversed without moving the probe to mitigate the effect of probe placement. The sample was then rotated 90 degrees and the resistivity was again measured for both polarities. The procedure was repeated twice more for a total of eight measurements. Because the forward polarity measurement is the same as the reverse polarity measurement on the sample rotated by 180 degrees an additional level of cross comparison was thus introduced. Figure 26 and Table 6 show the results.

![Effect of Probe Polarity on Measured Conductivity](image)

*Figure 26: Effect of Probe Polarity on Measured Conductivity*

*Table 6: Results of Probe Polarity Testing*

<table>
<thead>
<tr>
<th>Conductivity [W/m-K]</th>
<th>Positive</th>
<th>Negative</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rotation 0</td>
<td>23.4</td>
<td>26.9</td>
</tr>
<tr>
<td>Rotation 90</td>
<td>27.8</td>
<td>32.0</td>
</tr>
<tr>
<td>Rotation 180</td>
<td>29.9</td>
<td>44.5</td>
</tr>
<tr>
<td>Rotation -90</td>
<td>28.9</td>
<td>26.2</td>
</tr>
<tr>
<td>Mean (σ = standard deviation)</td>
<td>27.5 (2.9)</td>
<td>32.4 (8.5)</td>
</tr>
</tbody>
</table>
The direction of applied current was not found to impact the measured conductivity, as expected.

Size of Sample
To test the effect of sample size six cubes of side lengths from 10mm to 4mm in 1mm increments were fabricated from a single block of steel. The 10mm and 9mm cubes were used in the previous experiments. The resistivity of each sample was then measured to determine the thermal conductivity. Figure 27 and Table 7 show the results.

![Graph showing the effect of sample size on measured conductivity](image)

*Figure 27: Effect of Size of Sample on Measured Conductivity*

<table>
<thead>
<tr>
<th>Sample</th>
<th>Kappa</th>
<th>Kappa₀</th>
<th>RCF</th>
</tr>
</thead>
<tbody>
<tr>
<td>10mm³</td>
<td>36.5</td>
<td>38.0</td>
<td>0.9609</td>
</tr>
<tr>
<td>9mm³</td>
<td>26.1</td>
<td>27.6</td>
<td>0.9463</td>
</tr>
<tr>
<td>8mm³</td>
<td>27.8</td>
<td>29.9</td>
<td>0.9237</td>
</tr>
<tr>
<td>7mm³</td>
<td>26.0</td>
<td>29.3</td>
<td>0.8866</td>
</tr>
<tr>
<td>6mm³</td>
<td>25.7</td>
<td>31.3</td>
<td>0.8217</td>
</tr>
<tr>
<td>5mm³</td>
<td>32.3</td>
<td>46.1</td>
<td>0.7003</td>
</tr>
<tr>
<td>4mm³</td>
<td>24.1</td>
<td>52.3</td>
<td>0.4621</td>
</tr>
</tbody>
</table>
As can be seen from the results the thermal conductivity was approximately 28 W/m-K regardless of the sample size. This was not expected as the measured resistivity was expected to increase as the probe distance to the edge decreased with decreasing sample size. Further, application of the RCF was found to result in significant departure at small sample sizes. However, the results were consistent with the earlier sample geometry testing. An explanation for this is the high conductivity of the metal Aluminum.

Finally, the resistivity of an Aluminum sheet was measured over a range of 0 to 1A. Unlike the linear voltage current relationship expected and observed for steel the Aluminum was found to exhibit a non-linear voltage current relationship as shown in Figure 28 and the impact on measured resistivity can be seen in Figure 29.

![Figure 28: Measured voltage of Aluminum Sample Over a Range of 0-1A](image)

The measured resistivity of the aluminum sample was found to both depart significantly from the expected value and to be dependent on the applied current. Possible sources of error include contact resistance, sample geometry, and an incorrect reference value. Since aluminum metal readily forms an oxide layer the contact resistance may have been significant but is not accounted for. The aluminum sample was a thin sheet of thickness less than the probe separation distance.
Summary

The follow is a summary of the above experimental results. The voltage measurement was found to have greater consistency at higher applied current. The RCF was found to have little effect on calculated conductivity. The measured voltage was found to be sensitive to small electrical noise and was inconsistent at 10mA applied current. The thermal conductivity measurement was found to be repeatable at 80mA applied current. The probe polarity was not found to have a significant effect on measured conductivity. The technique was found to be insensitive to the size of the sample. It was found oxide layers can significantly affect the measured resistivity. The four-point probe was also found to be insensitive to applied pressure. The manufacture specifies that the pins should retract half way into the probe and it was observed empirically that no measurement can be made if the pin pressure is insufficient. The pin pressure is either sufficient or no measurement can be made.

Chapter 4: Experimental Results – AC Measurement

In an attempt to improve on the results from the DC experimentation a lock-in amplifier was used with an AC excitation current. In a lock-in amplifier, a reference signal of the same frequency as the measured signal is multiplied and integrated over time. The multiplication eliminates the noise that is of a different frequency from the signal. A function generator is used to create a sine wave which is sent to
both the sensor and the lock-in amplifier. The signal is distorted in the sensor and arrives at the lock-in
with noise. The multiplication and filtering of the signal results in a single DC voltage output which can
then be recorded by a multi-meter. First the probes response to applied frequency was investigated.
Next, the effect of applied current, sample size, and the repeatability was characterized the same as it
was for DC.

AC Current
The 9mm³ sample was measured at 1, 5, 10, 40, and 80mA applied currents at frequencies from
100-1000Hz. Figure 30 shows the voltage is dependent on both applied current and frequency.

![Voltage Response to AC Frequency](image)

*Figure 30: Voltage Response on AC Frequency and Current*

Based on the results from the DC testing the voltage response is not expected to depend on applied
current. Figure 31 plots the voltage over current dependence on frequency. As can be seen the current
dependence is removed and an approximately linear dependence on frequency is evident. Applying a
linear curve fit and extrapolating to the equivalent DC response at zero frequency the value for V/I was
found to be 32.45 µV mA which corresponds to a thermal conductivity of 29.7 W m⁻¹ K⁻¹.
Applied Frequency

The observed frequency dependence is likely a result of self-inductance within the material as self-inductance is proportional to frequency and would not be observed for DC measurements.

Modeling the material between the pins of the four-point probe as a resistor and inductor:

\[ V_R = \frac{\rho}{2\pi s} I \rightarrow \frac{V}{i} = R \]  \hspace{1cm} (29)

**Self-inductance**\[ V_L = 2\pi L f I \rightarrow \frac{V}{i} = X_L \]  \hspace{1cm} (30)

Modeling the resistance and reluctance to be in series the impedance is:

\[ Z = \sqrt{X_L^2 + R^2} \rightarrow Z = \sqrt{(2\pi L f)^2 + \left(\frac{\rho}{2\pi s}\right)^2} \]  \hspace{1cm} (31)

Therefore, the relationship between \( \frac{V}{I} \) and frequency is:

\[ \frac{V}{I} = \sqrt{(2\pi L f)^2 + \left(\frac{\rho}{2\pi s}\right)^2} \]  \hspace{1cm} (32)

Which reduces to the familiar \( \frac{V}{I} \) relationship for DC at zero frequency. Measurements taken over a wide range of frequencies were curve fit both with a linear trend and with the relationship above and is plotted in Figure 32 below.
The coefficient on the frequency term for the line of best fit was 0.35 H. Taking inductance of the material to be equal to the permeability times the probe separation distance the reluctance of the material was calculated to be 40 $\mu$H/m. For comparison the permeability of free space is 1.24 $\mu$H/m and of pure iron is 125 $\mu$H/m with 40 $\mu$H/m within the expected range for steel. Several studies investigating the use of the four-point probe with AC also reported an inductive effect and successfully measured electrical resistivity [25] [31] [32]. The formulation for the voltage in the material accounting for inductive effects is reproduced in equation 33 below. However, the data collected in this study was not successfully applied to the model.

\[
V = \frac{I}{\tau} \left[ -\frac{1}{\tau} + \mu_0 \mu_0 \left( \frac{\mu T}{3} + l \right) - \frac{\omega^2 \mu^2 c T^3}{45} + O(\omega^2) \right] \times \ln \left| \frac{1 + a}{1 - a} \right|, \quad f < f_v \tag{33}
\]

Only the linear model was found to fit the data collected in this study, the remained results are reported based on linear extrapolation from measurements taken at three different frequencies to
remove the frequency dependence of the voltage signal. Based on the performance on the linear model at high and low frequency ranges the optimal frequency was determined to be 200-300 Hz.

**Applied Current**

Figure 33 shows the current dependence of the measured thermal conductivity. The measurements were taken at 250 Hz on the 9mm$^3$ sample at currents from 0.17 to 92.5 mA.

![AC Current Dependence of Kappa](image)

*Figure 33: AC Current Dependence of Measured Thermal Conductivity*

A current dependence was observed at low currents, and a stabilization at high currents, as observed for DC. High applied current should be avoided to mitigate damage to the probe and current saturation. 10 mA was found to optimally balance low and high current effects.

**Repeatability**

To test the repeatability of the measurement using AC the 9mm$^3$ sample was measured on all six sides at 0 and 90 degrees at 9.37 and 11.12 mA. Figure 34 depicts the results plotted with both the linear and non-linear models. Table 8: Results of Repeatability Testing with AC Linear Curve FitTable 8 tabulates the results.
The thermal conductivity was measured to be 31 W/m-K with a comparable amount of variation to the DC measurements. As can be seen from Figure 34, the non-linear model had less spread in the calculated thermal conductivity but was between 15-20 W/m-K. The frequency dependent term in the non-linear model dominates resulting in decreased variation. Figure 35 compares the repeatability testing of both DC and AC measurements. The repeatability at 10 mA AC and 80 mA DC were comparable and better than the repeatability at 10 mA AC. Therefore, by taking measurements with AC less current needs to be applied. This is beneficial because the applied current should be minimized.
Sample Size

Finally, the effect of sample sized was tested by measuring samples of decreasing side lengths at 9.37 and 11.12 mA. The results are plotted in Figure 36.
The results were the same as for the DC case. The measured thermal conductivity did not depend on the size of the sample over the range tested and applying the RCF resulted in erroneous values for thermal conductivity.

Chapter 5: Conclusions.

The goal of this study was to develop a reliable and simple method of measuring thermal conductivity in metal fuels using electrical contact or show comprehensively the prohibitive limitation of this approach. The four-point probe and apparatus used in this study was simple and easy to use. The approach was found to be insensitive to inexact probe placement, applied probe pressure, temperature, and radiation environments. The specific outcome goals of this research were:

- Insuring good pin contact on material substrate

The pin pressure is either sufficient or no measurement can be made. For the apparatus used increasing the weight on the probe head was found to insure adequate pressure.

- Accurate Resistivity Correction Factors to determine electrical resistivity

A sensitivity analysis of RCF for different geometries was conducted and suggested the probe is insensitive to inexact placement. Further, the experimental results suggest that the RCF’s are not needed for samples of the size tested.

- Accurate model to relate conductivities based on the Wiedemann Franz Law

The Wiedemann Franz Law is a well-established method of relating electrical and thermal conductivity. Published results suggest the Sommerfeld value of the Lorenz number is not expressly correct but is an approximation that can be used if no other material properties are known.

- Measurement tool and apparatus applicable to use in hot cell
The apparatus used to hold the probe and the samples would be easy to implement in a hot cell and would be easy to operate. A conceptual model of a potential implementation is shown in Figure 37.

![Conceptual Apparatus Design for Cylindrical Samples in a Hot Cell](image)

Figure 37: Conceptual Apparatus Design for Cylindrical Samples in a Hot Cell

One limitation of using the four-point to determine the thermal conductivity is that the Weidemann-Franz can only be used to relate the electrical conductivity and the thermal conductivity due to electrons. In order to use the four-point probe for thermal conductivity measurements it must be assumed that phonon thermal conduction is not significant. This assumption is generally valid for metals. However, the four-point probe is not well suited for materials with high thermal conductivity, such as metals, because of the low voltage gradient in the material under the applied current therefore resulting in poor resolution for metals.

Given additional time and resources, the results of this study suggest several areas of future work. Characterizing the effect of surface roughness on measured thermal conductivity was not tested in this study, however, empirical observations during repeatability testing suggests contact resistance on rough samples may be significant. Additionally, validation of the technique against a sample of known electrical or thermal conductivity would be beneficial as would the measurement of a novel material of
unknown material properties. The comparison between the DC and AC measurement results suggest that future work should be focused on AC. In particular a model providing a more robust explanation of the observed frequency dependence. Such a model was reported by Bowler [32], however, the data collected in this study did not fit the model. Another significant area of future work is the RCF for extremely thin or small samples. The resistivity of a few nanometer gold film was measured with the apparatus but the RCF was found to be insufficient for such a thin sample. Finally, the use of eddy currents for electrical conductivity measurement has been investigated [26] and could be of significant practical value for sub-surface thermal conductivity measurements.

In conclusion, the use of the four-point probe for measurements of nuclear fuels in hot cells is feasible, easily implementable, and has a resolution of about 5 W/m-K. Further research could likely increase the accuracy and resolution of the method without compromising the easy and simplicity of using the technique on metallic nuclear fuels.
References


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